Signatures of Random Matrix Theory in the Discrete Energy Spectra of Shape Disordered Metallic Clusters

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Brian Lang

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16 November 2005

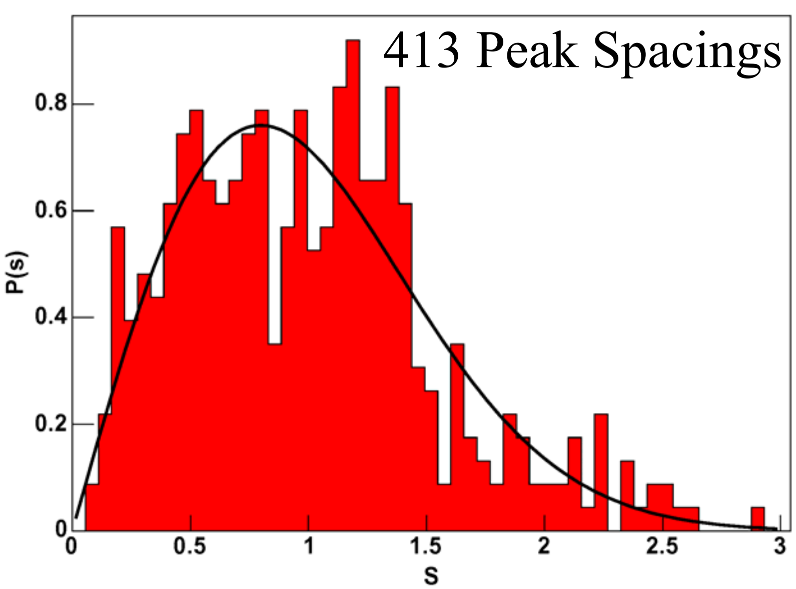
Preview

It is expected that the statistics of the discrete energy spectra of small disordered metal grains should be described by random matrix theory (RMT).[1]

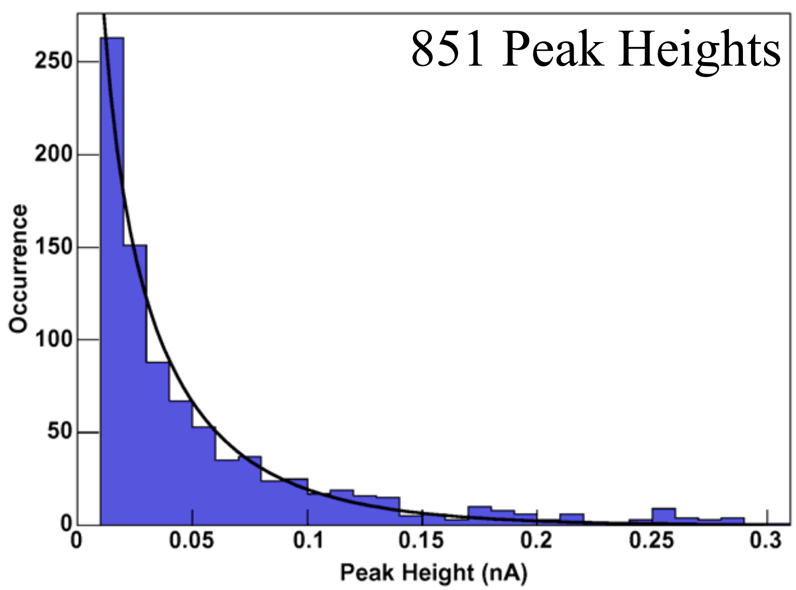
Narvaez and Kirczenow, PRB **65**, 121403 (2002).

M. L. Mehta, Random Matrices (Academic Press, New York (1967)); W. P. Halperin, Rev. Mod. Phys. **58**, 533 (1986); E. B. Efetov, Adv. Phys. **32**, 53 (1983); T. A. Brody *et al*., Rev. Mod. Phys. **53**, 385 (1981); A. D. Mirlin, Phys. Rev. **326**, 360 (2000); Y. Alhassid, Rev. Mod. Phys. **72**, 895 (2000).

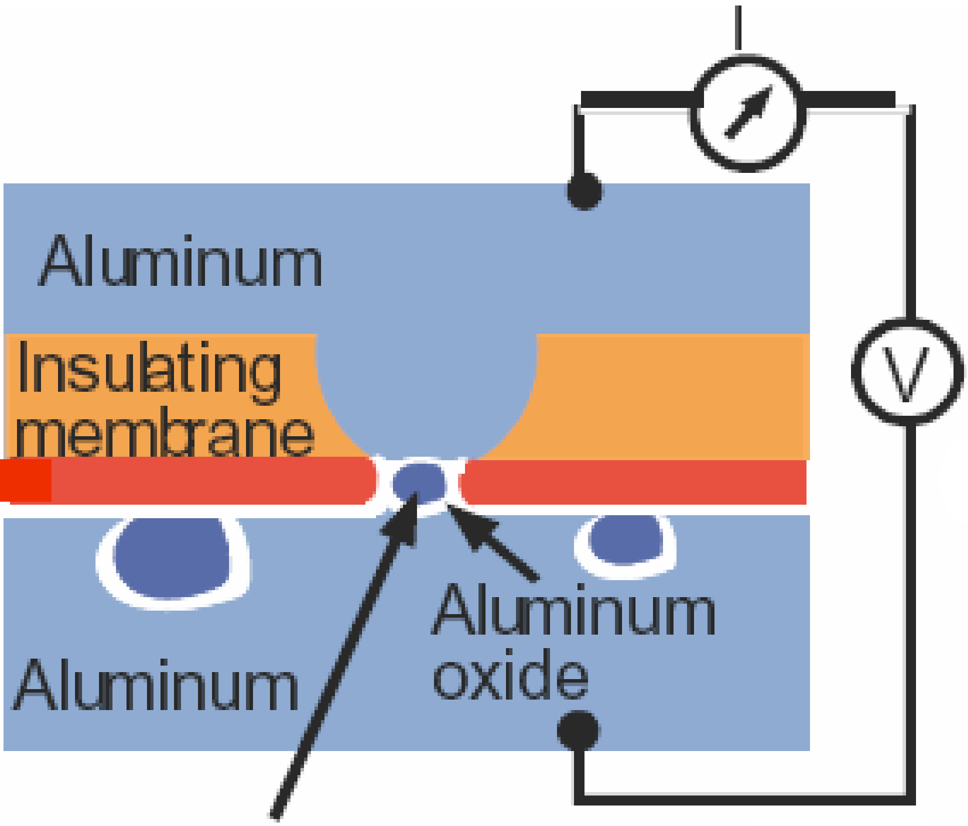
Wigner-Dyson Distribution



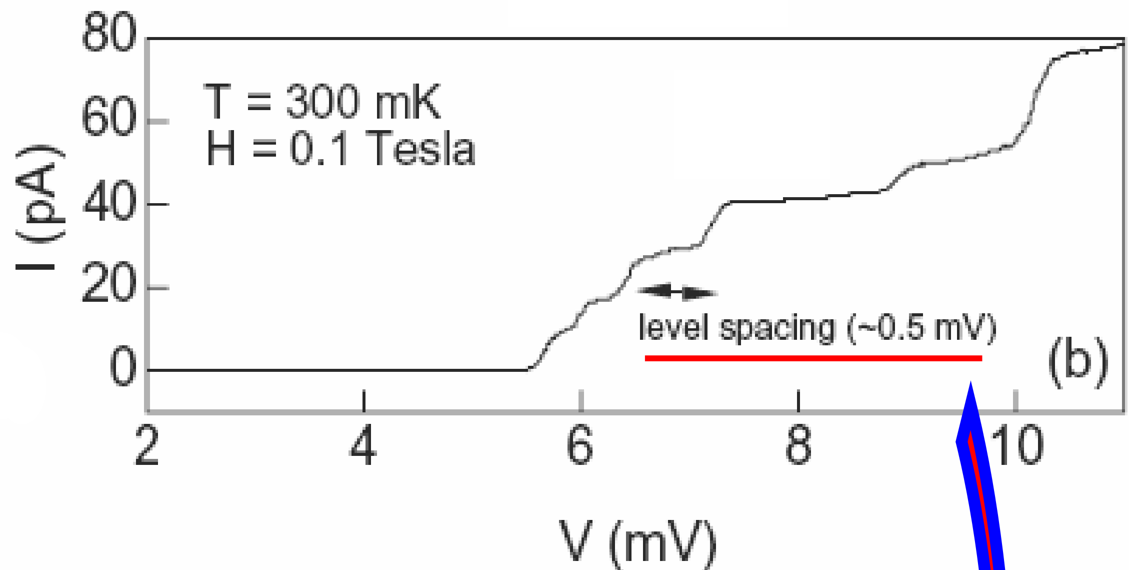
Porter-Thomas Distribution



First Observation of Level Spacings in Metallic Clusters



Ralph *et al.*'s Pioneering Experiments



Ralph, Black and Tinkham, PRL **74**, 3241 (1995).

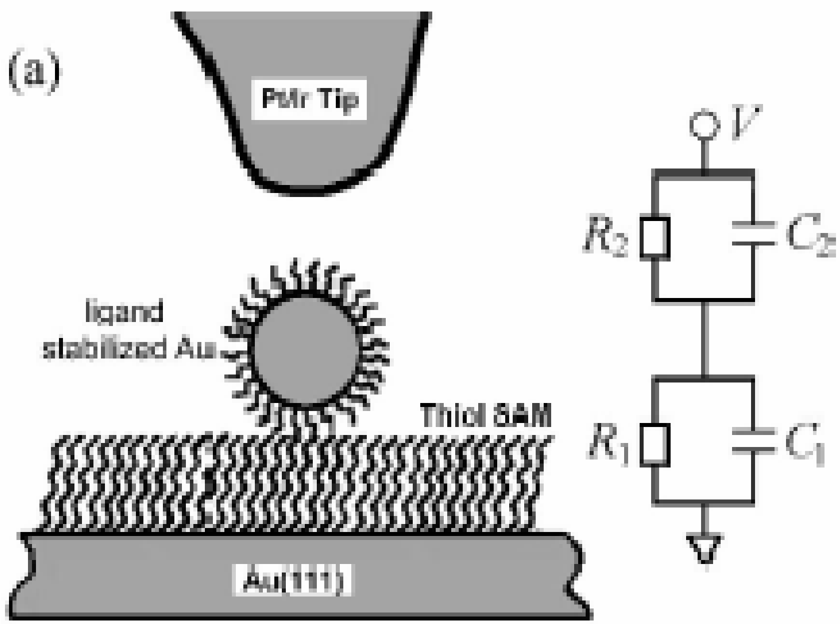
Mean Level Spacing, Δ:

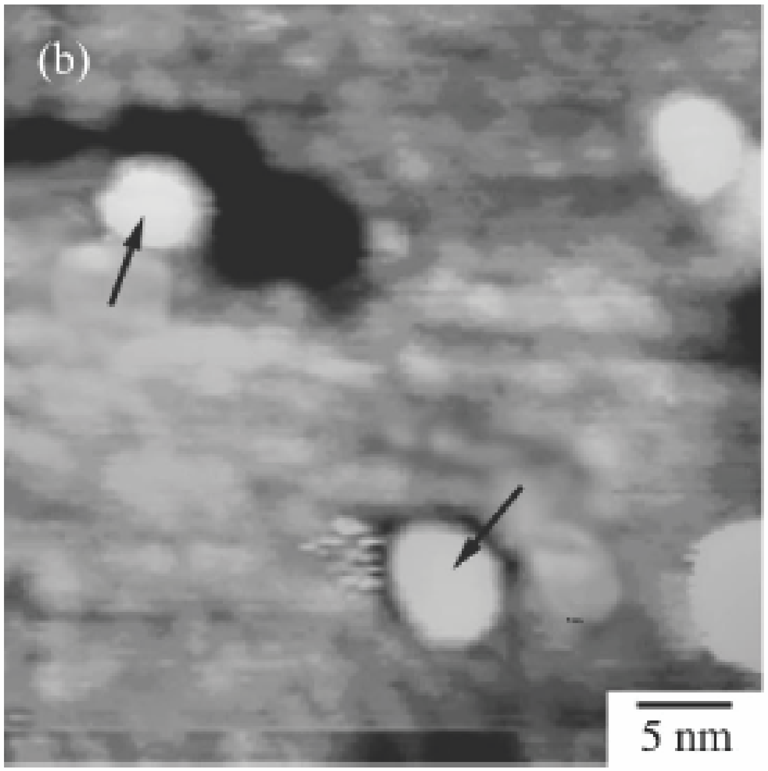
$$\boxed{\Delta_{3D} = \frac{1}{DOS} = \frac{2\,\pi^2\,\hbar^2}{m\,k_f\,\mathrm{Vol}}}$$

Δ_{3D} (theoretical) = 0.3 meV for a cluster of radius 5 nm.

Another Approach to Resolving Discrete Energy Levels

Scanning Tunneling Microscope Image

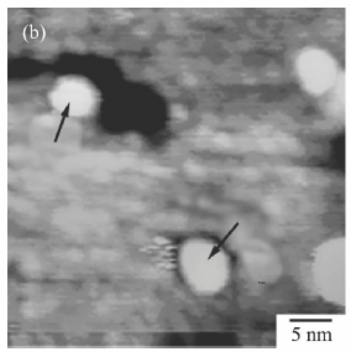




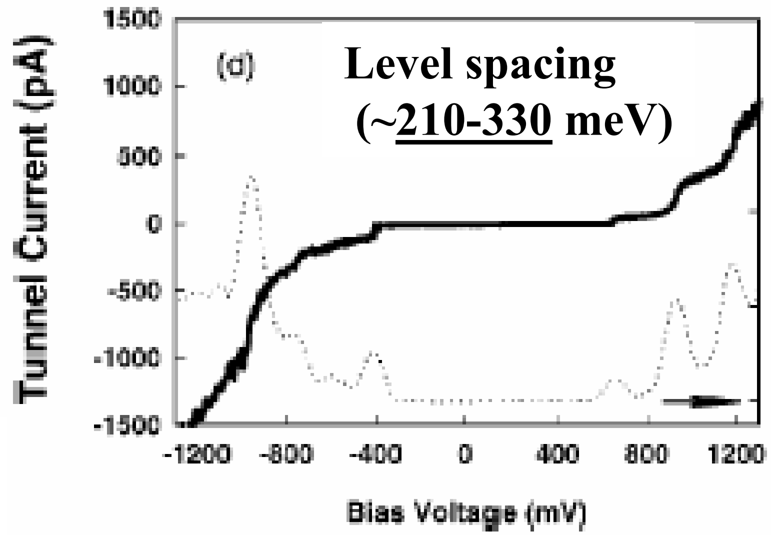
Wang *et al.*, PRB **63** 035403 (2000).

Similar Experimental Results: Irregularly Spaced Current Steps

Data Taken at 4.2K



$$\Delta_{3D} = \frac{1}{DOS} = \frac{2\,\pi^2\,\hbar^2}{m\,k_f\,\mathrm{Vol}}$$



Cluster Size

18 Å

Wang *et al.*, PRB **63,** 035403 (2000)

Cluster Diameter	Δ_{3D} (theoretical)
10 Å	~ 240 meV
15 Å	~ 70 meV

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Some Questions about Discrete Energy Levels in Metallic Clusters

- Are the uneven level spacings a signature of **random matrix theory**?

- Is it **experimentally** possible to acquire a sufficient number of levels to generate statistical distributions?

- How does the **shape** of the clusters effect the **distribution** of energy levels?

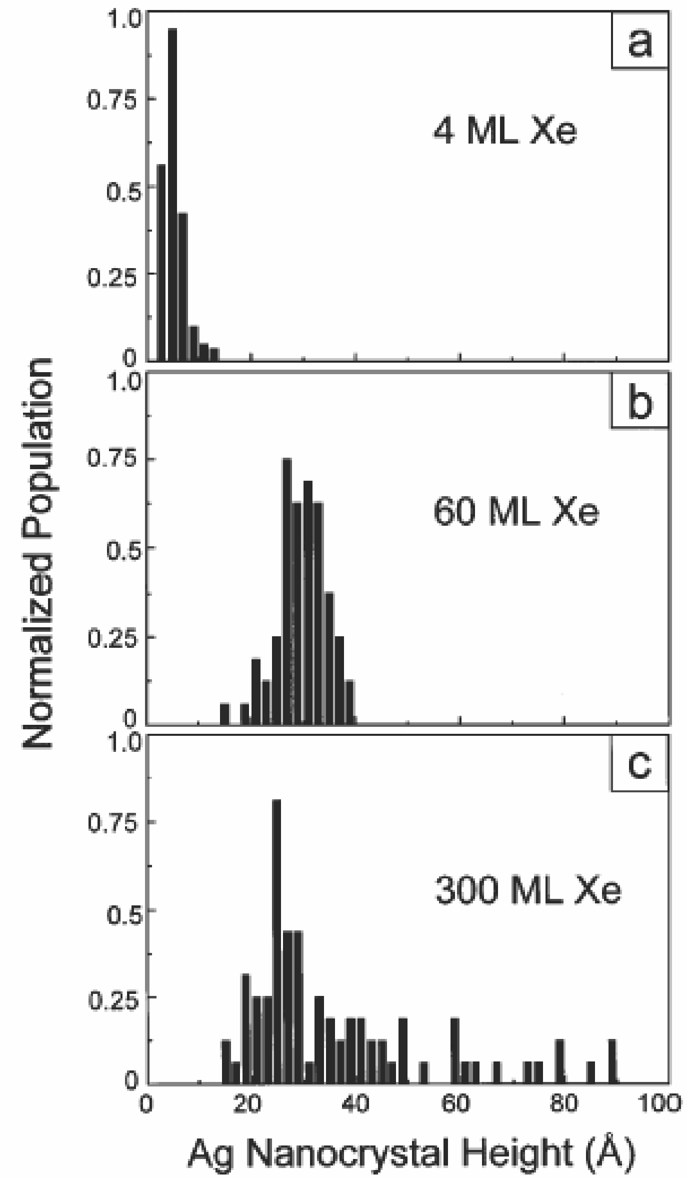
Overview of our experiment

1. Fabricate nanometer size clusters *in situ* using a buffer layer assisted growth technique.

(Technique developed by **John Weaver and collaborators**.)

2. Measure the size of the clusters using a Scanning Tunneling Microscope.

3. Measure the current-voltage characteristics through the cluster using Scanning Tunneling Spectroscopy at liquid helium temperatures.



Huang, Chey and Weaver, PRL **80**, 4095 (1998)

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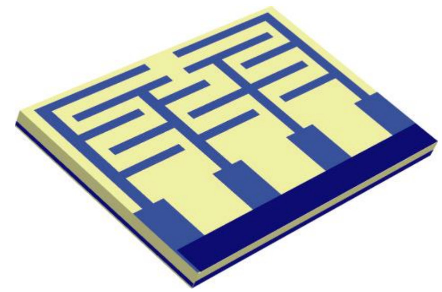
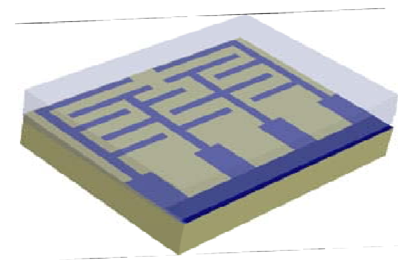
Fabrication of Nanosize Pb Clusters: Buffer Layer Assisted Growth Technique

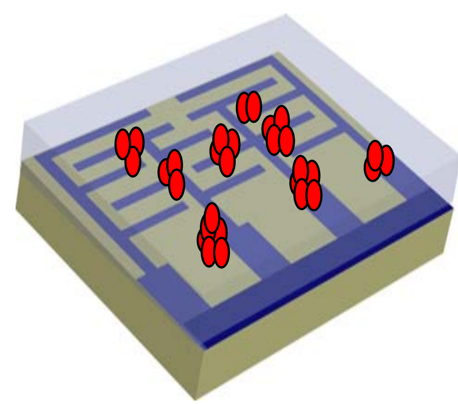
Deposit Pt/Ti electrodes onto Si (111) $\rho \geq 1000\ \Omega$ cm at room temperature.



Cool to T < 50 K and deposit 4 monolayers of Xe on top.



Deposit a fraction of a monolayer of material on top of the Xe at T < 50 K.



Gradually warm to room temp. which allows the clusters to land softly onto the substrate.

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Huang, Chey and Weaver
PRL 80, 4095 (1998)

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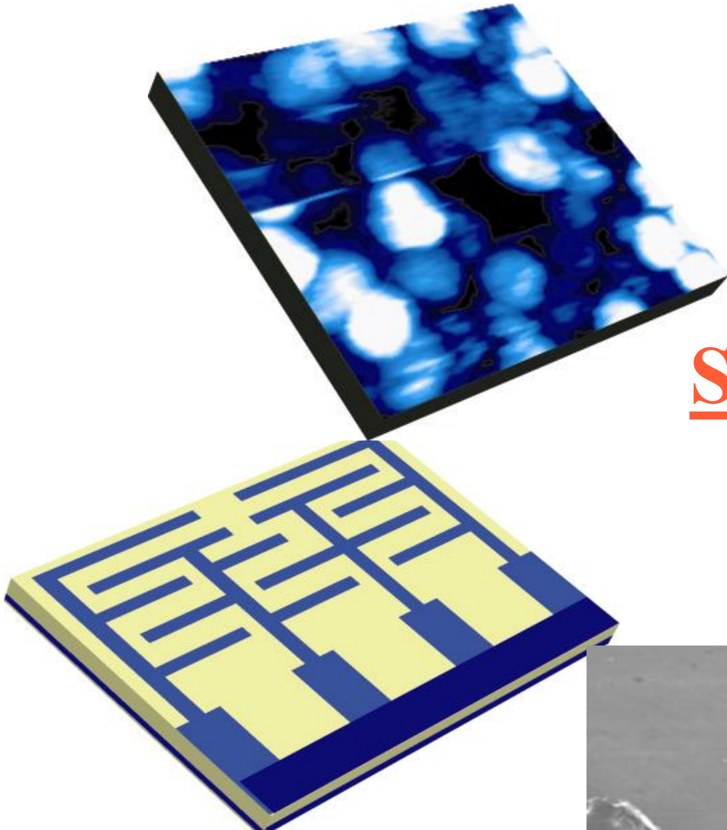
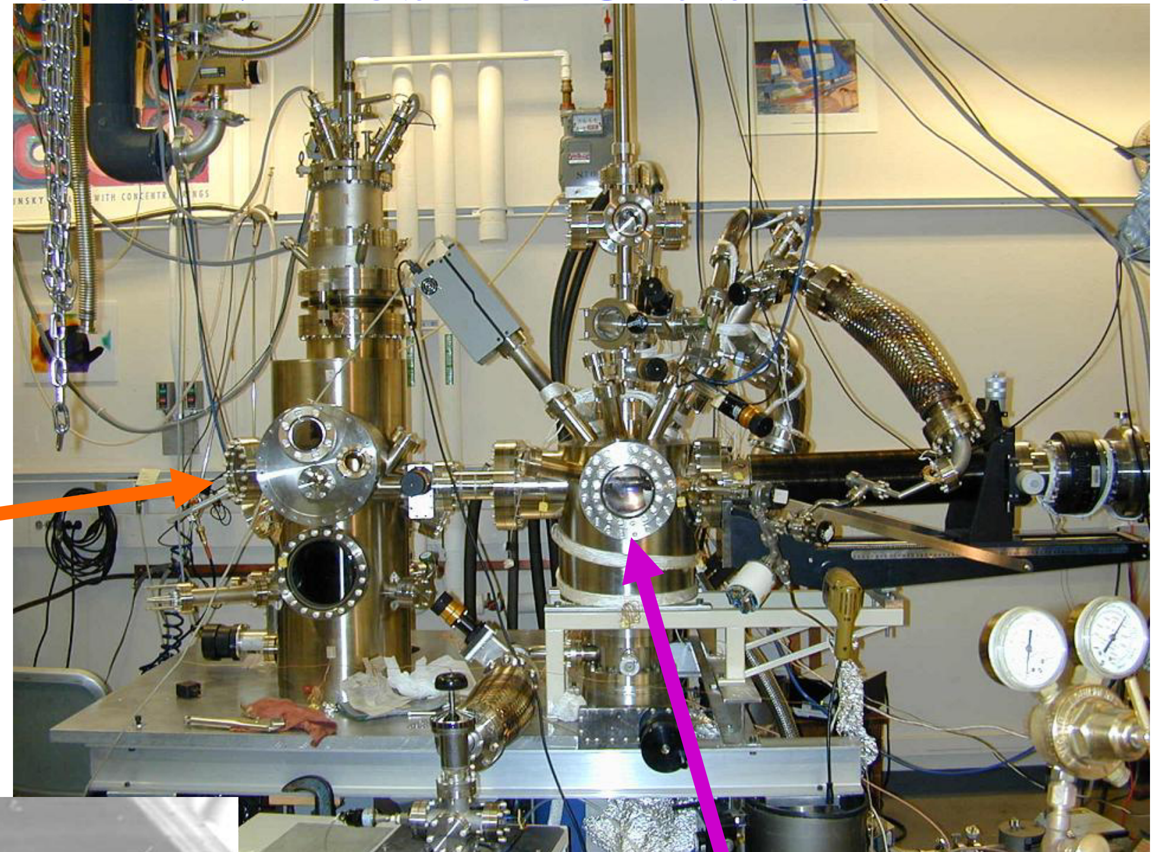
Measurement of Lead Nanosize Clusters

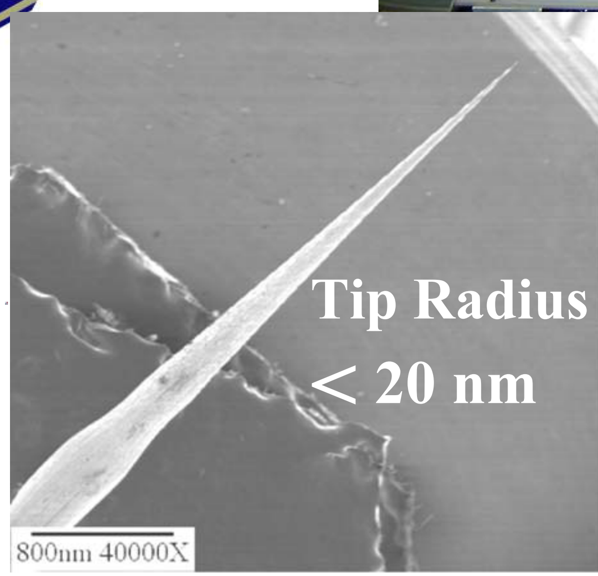
Soft Landing Technique





STM

Deposition Chamber



L. L. A. Adams and A. M. Goldman, RSI **76**, 063907 (2005).

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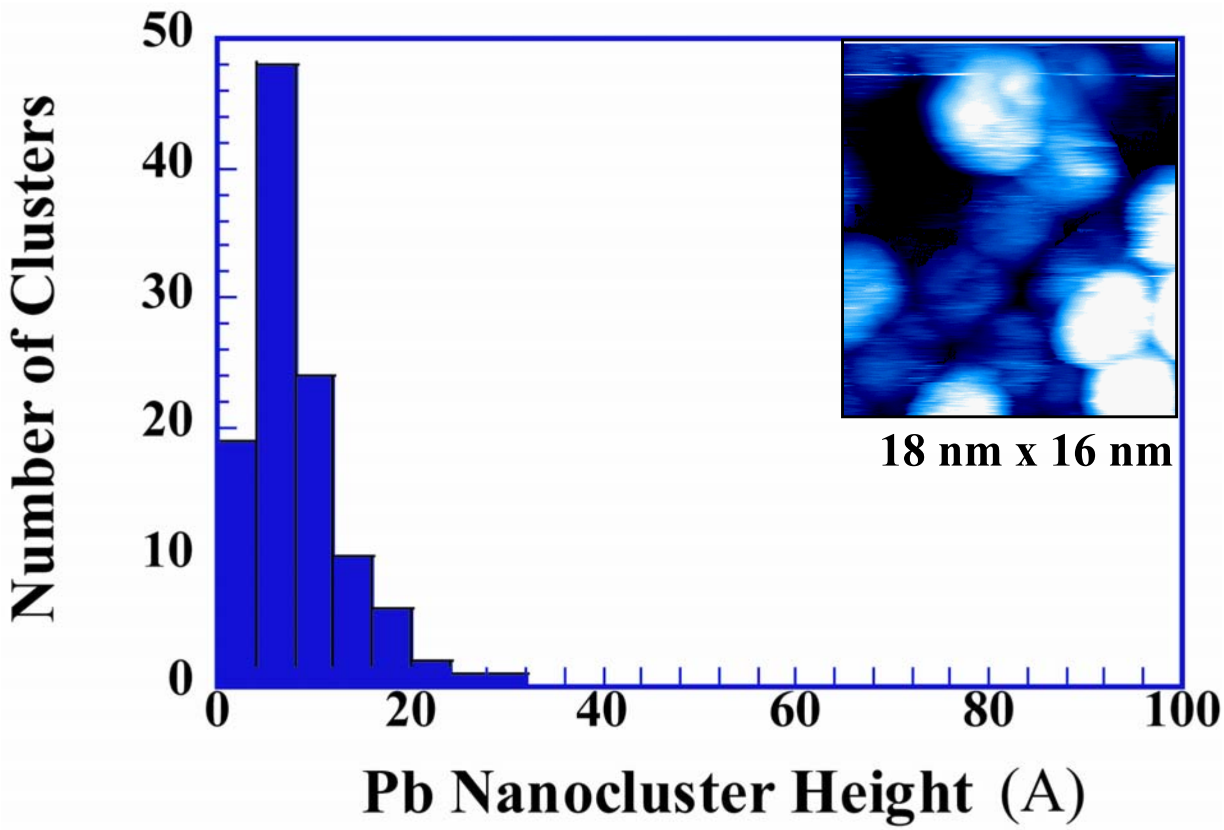
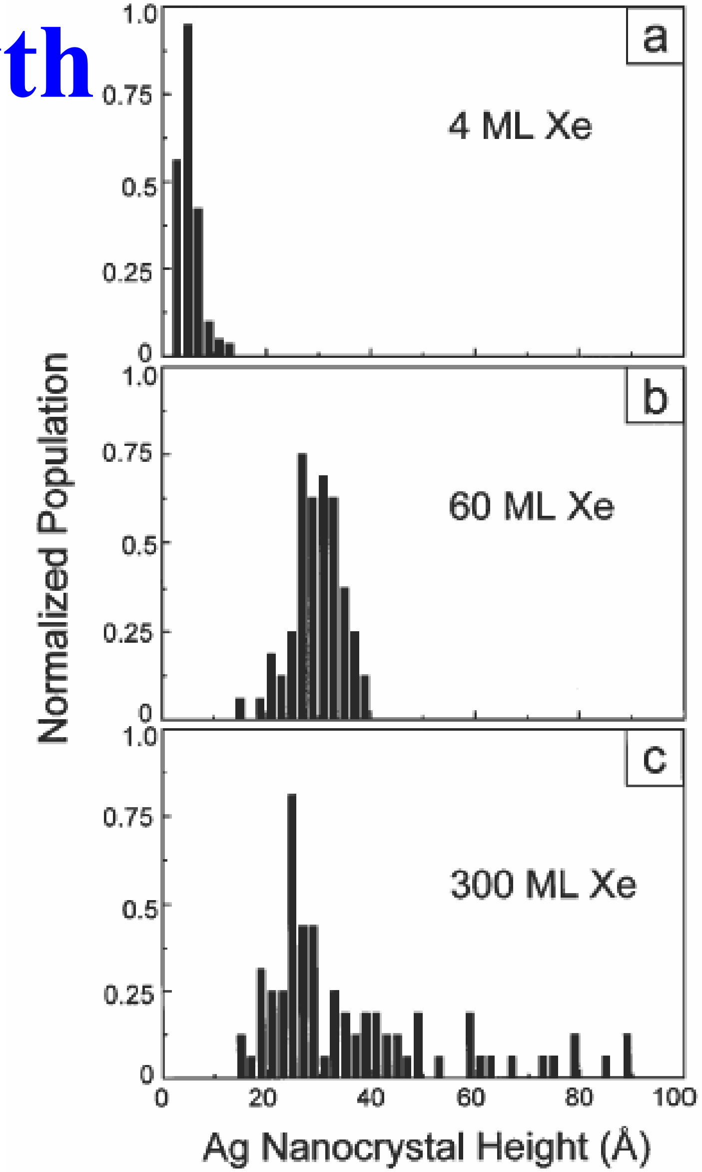
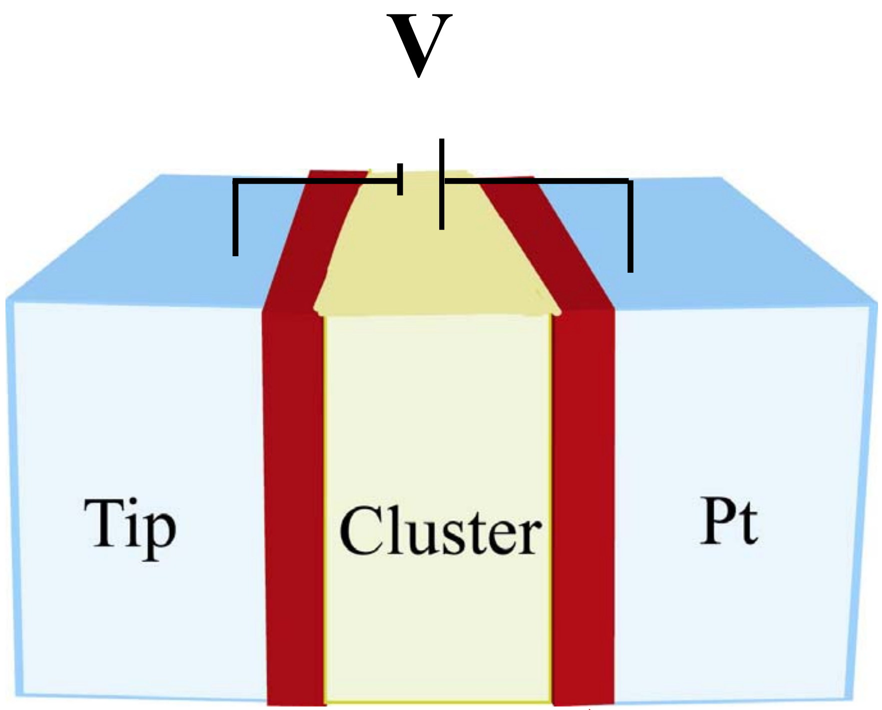
Expected Results: Pb Cluster Growth





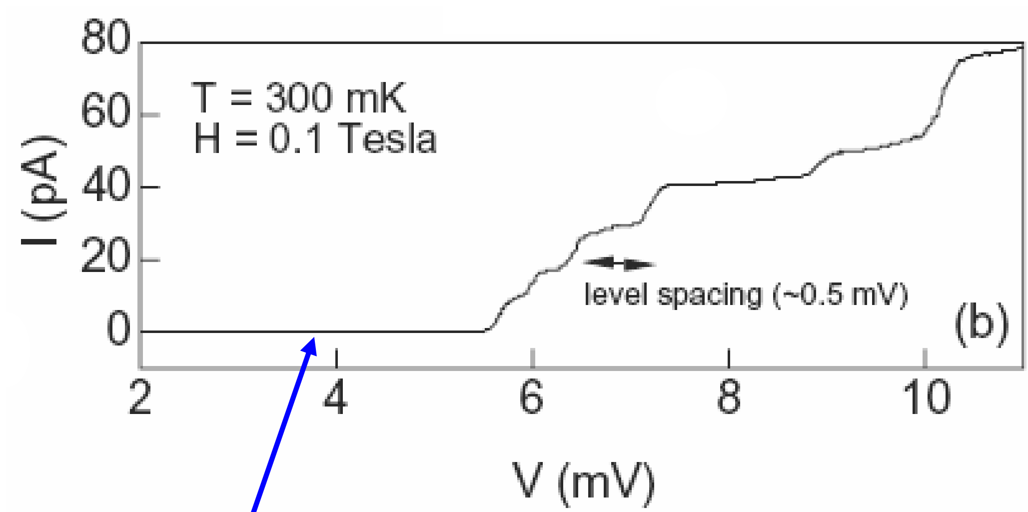
Huang, Chey and Weaver, PRL **80**, 4095 (1998)

What was expected



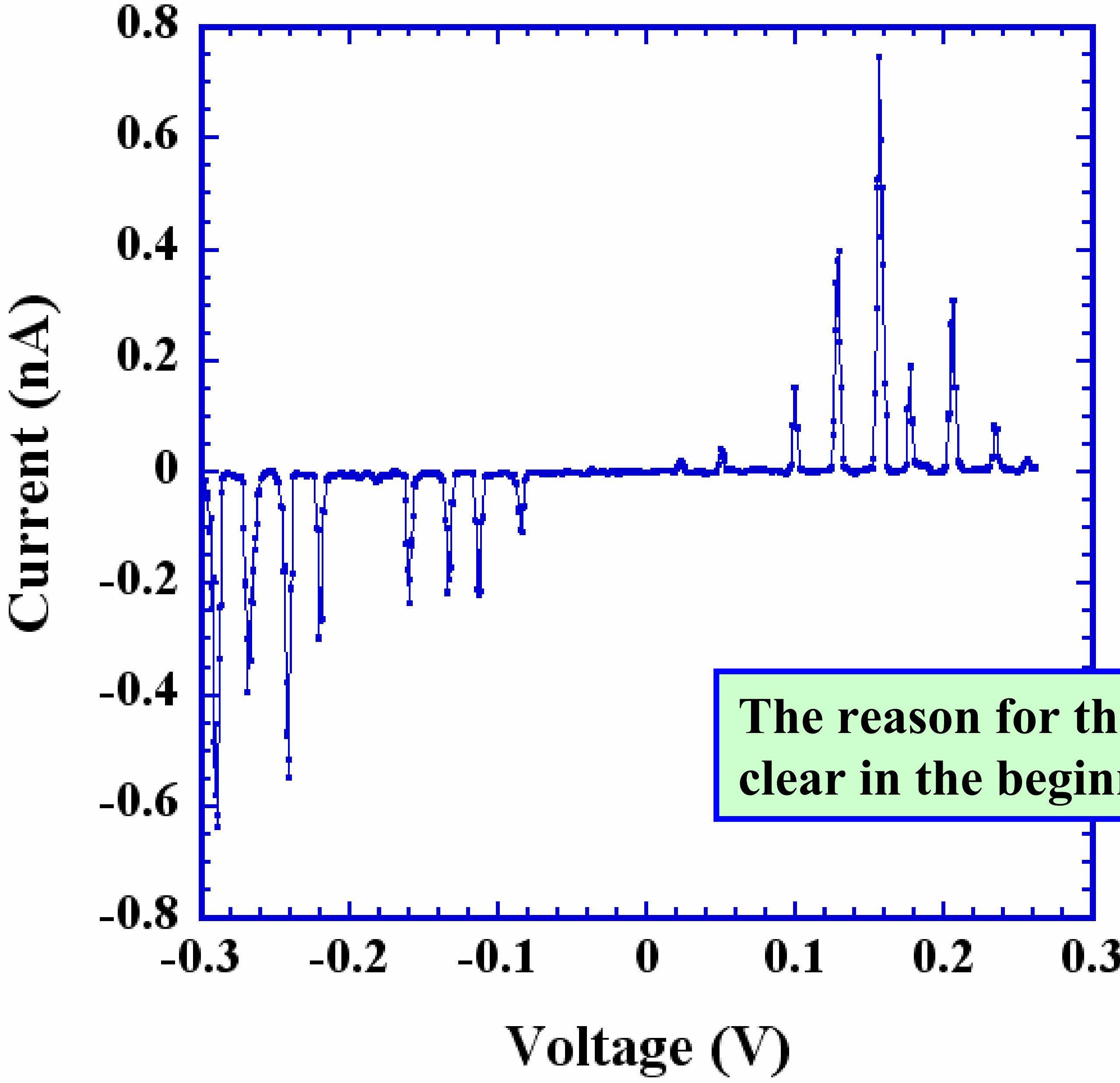
Vacuum Barrier

Silicon Barrier
(Insulating at 4 K ?)



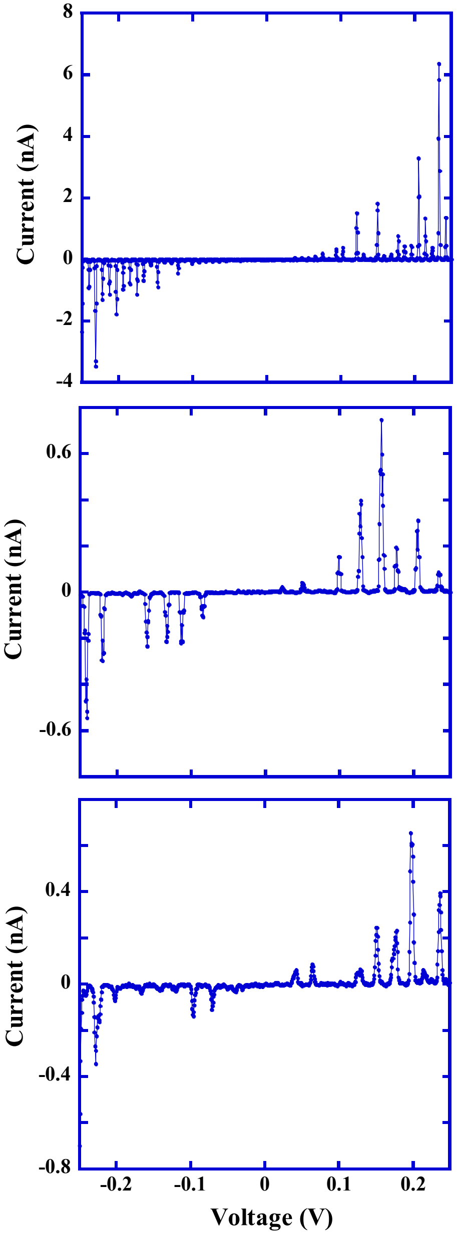
Coulomb Blockade

What was observed….

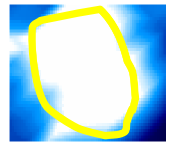


The reason for the peaks was not clear in the beginning…

Size Matters: Mean Peak Spacing as a function of Cluster Size



Cluster Size
Length = 4.3 nm
Width = 1.5 nm
Height = 2.2 nm

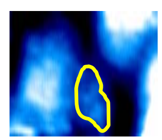


Mean Peak Spacing ~ 9.6 meV
Δ (estimated) ~ 2 meV

Cluster Size
Length = 2.7 nm
Width = 2.97 nm
Height = 0.6 nm

Mean Peak Spacing ~ 10.8 meV
Δ (estimated) ~ 6 meV

Cluster Size
Length = 2.5 nm
Width = 1.7 nm
Height = 0.34 nm



Mean Peak Spacing ~ 24.2 meV
Δ (estimated) ~ 20.3 meV

Decreasing Cluster Size

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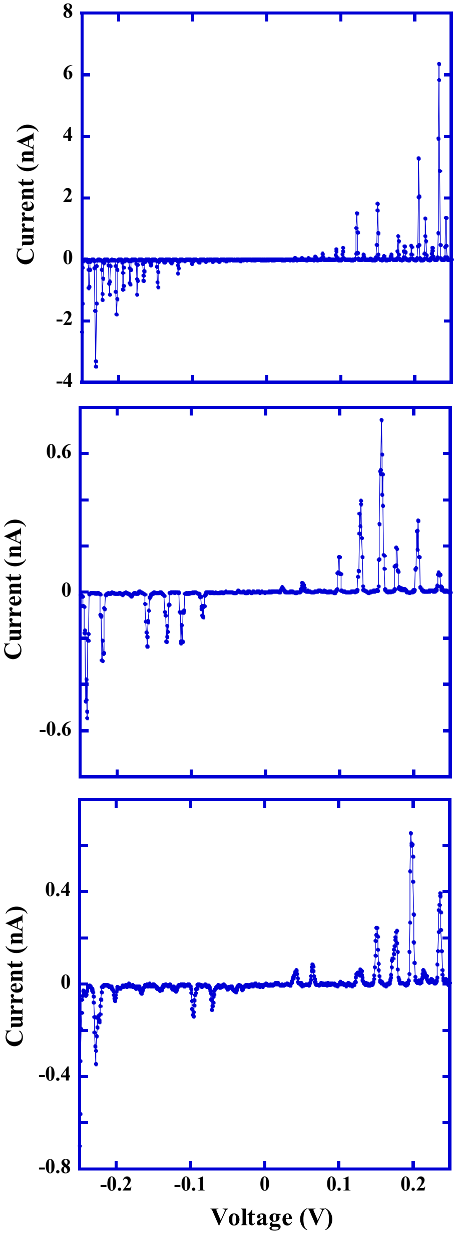
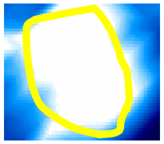
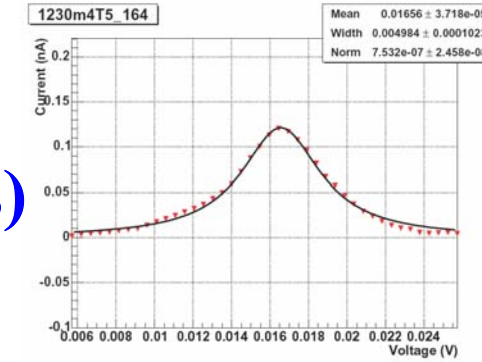
Size Matters: Peak Linewidth as a function of Cluster Size

Cluster Size
Length = 4.3 nm
Width = 1.5 nm
Height = 2.2 nm





Linewidth ~ 1.06 meV
Tunnel time: 6.2×10^{-13} seconds



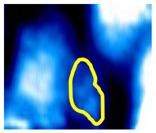
(Lorentzians)

Decreasing Cluster Size

Cluster Size
Length = 2.7 nm
Width = 2.97 nm
Height = 0.6 nm

Linewidth ~ 3.06 meV
Tunnel time: 2.4×10^{-13} seconds

Cluster Size
Length = 2.5 nm
Width = 1.7 nm
Height = 0.34 nm



Linewidth ~ 3.54 meV
Tunnel time: 1.9×10^{-13} seconds

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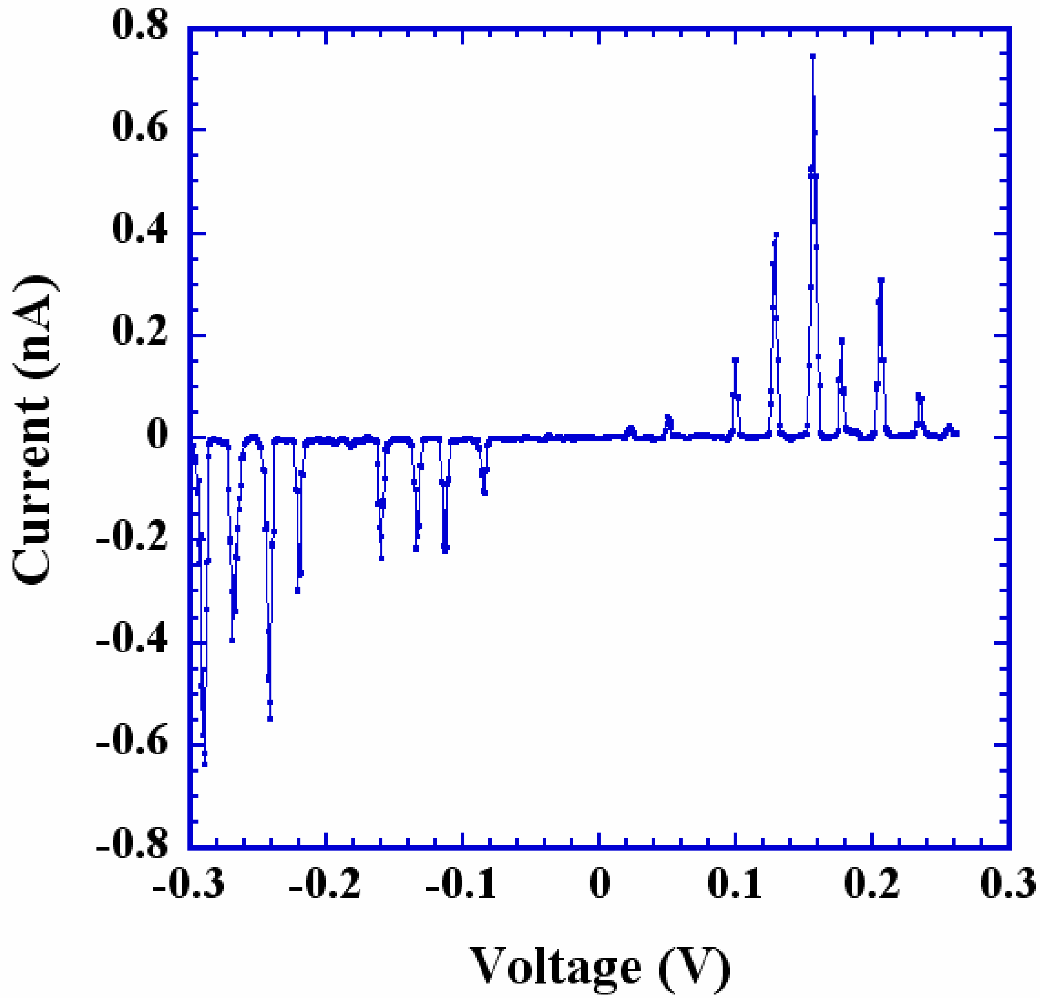
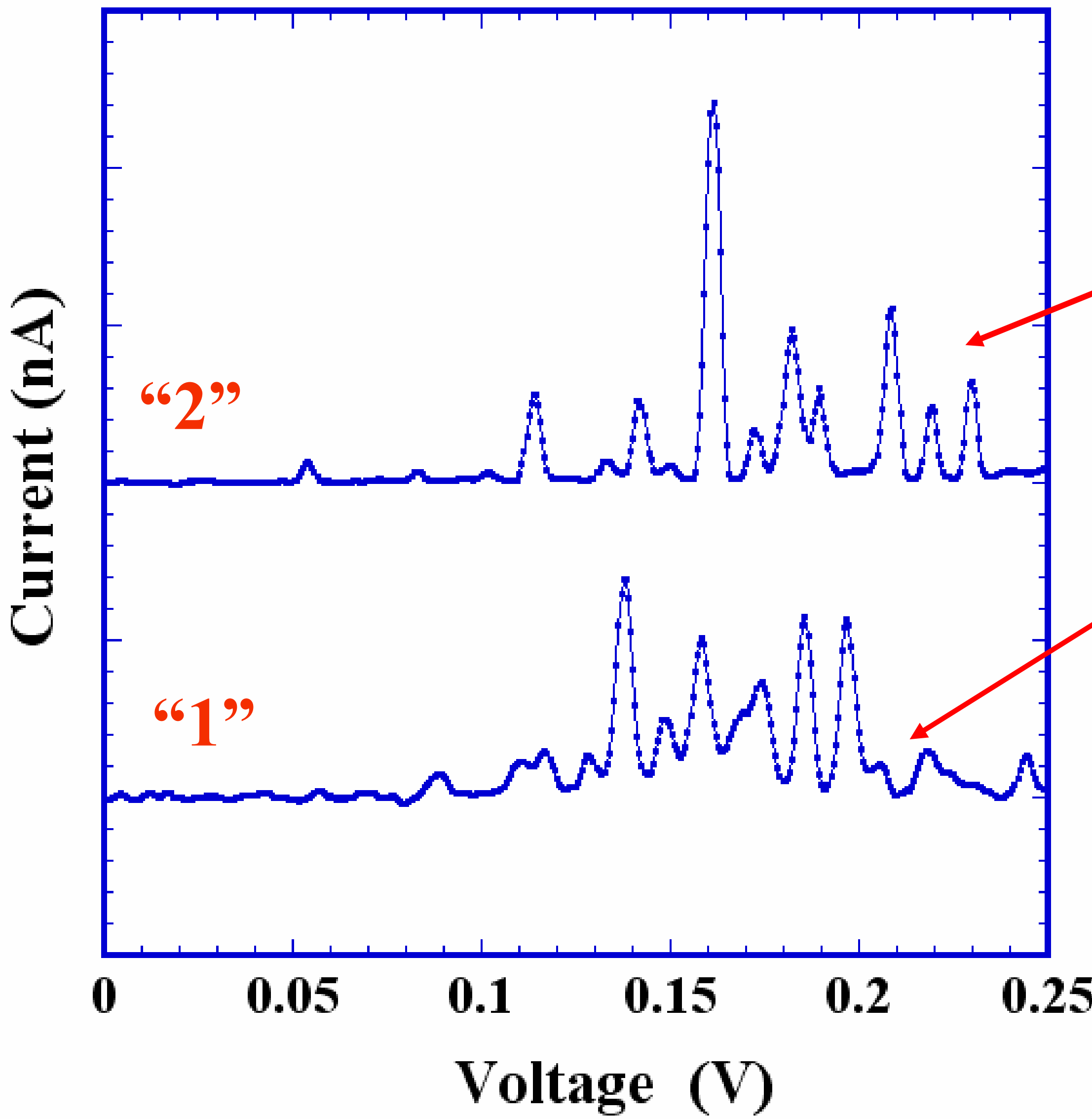
Interesting Aspects of the Data

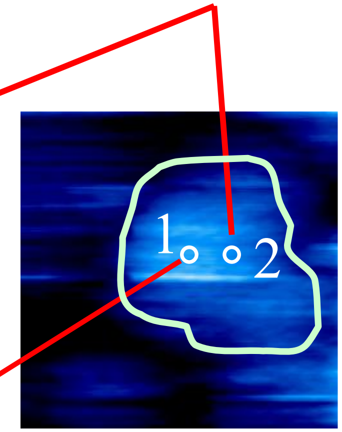


- **Peak spacings are *not* equally spaced.**
- **Peaks (and the spacing between the peaks) varies as a function of position along a cluster.**

Cluster's dimensions are:
length ~ 3.7 nm, width ~ 2.6 nm, height ~ 0.3 nm

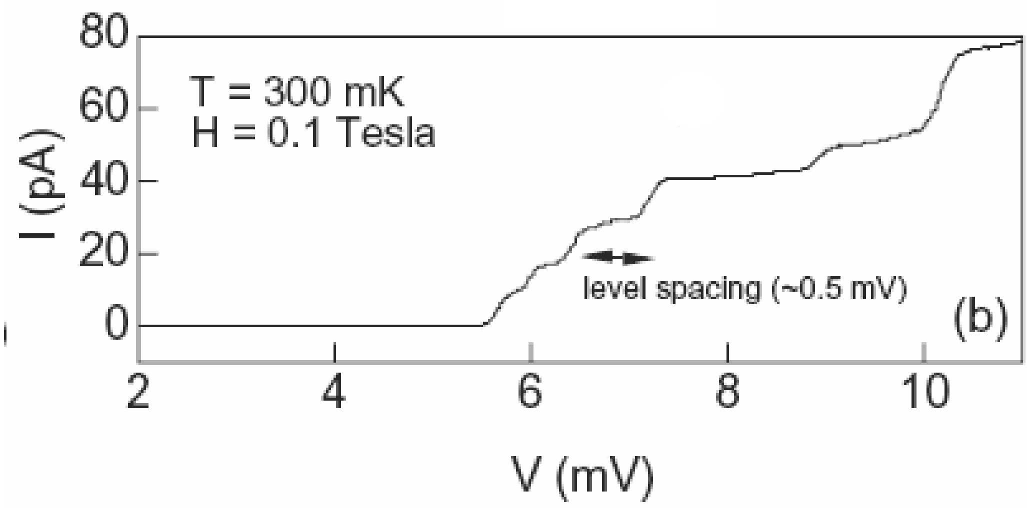
Position Dependent Current-Voltage Characteristics

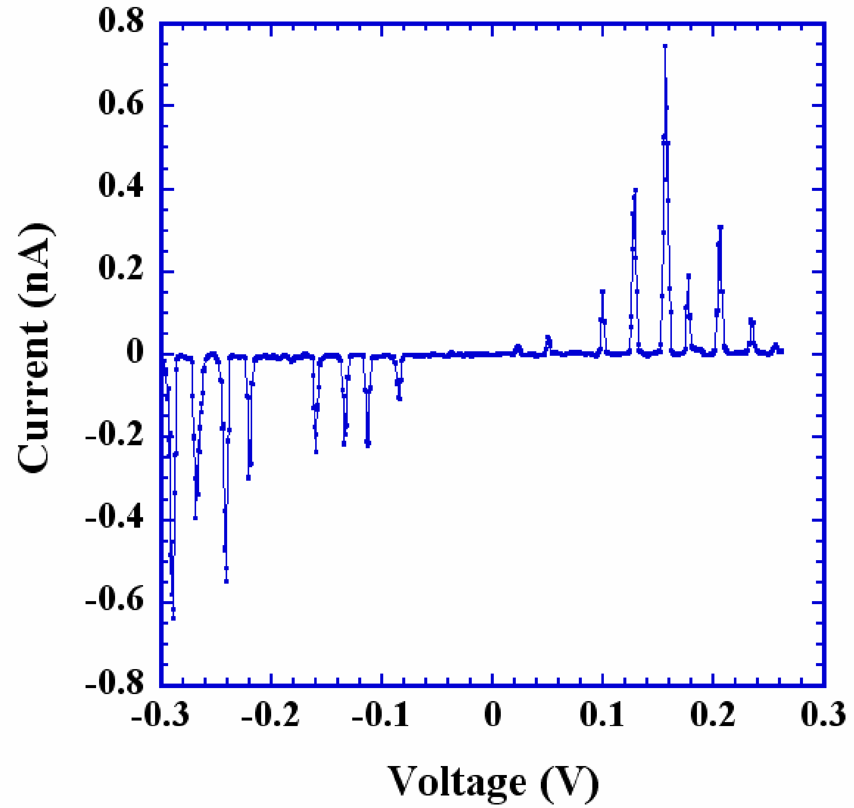


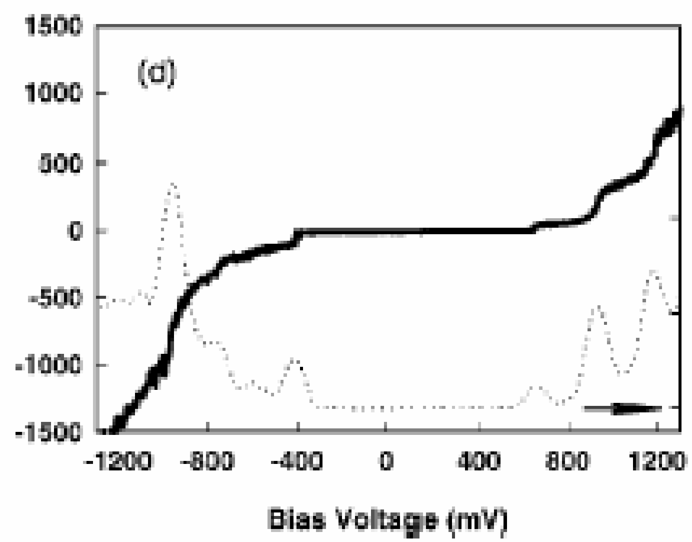


Cluster Diameter: ≈ 36 Å
Cluster Height: ≈ 3 Å

Differences between experimental data on metallic clusters







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Differences between this experiment in comparison to others

Three Important Differences

1.) Used a **semiconducting** substrate instead of a metallic substrate.

2.) Silicon substrate was **highly resistive**.
Si(111) substrate
Phosphorous-doped
Resistivity > 1000 Ohm-cm

3.) **Soft landing technique** to deliver the clusters to the substrate.

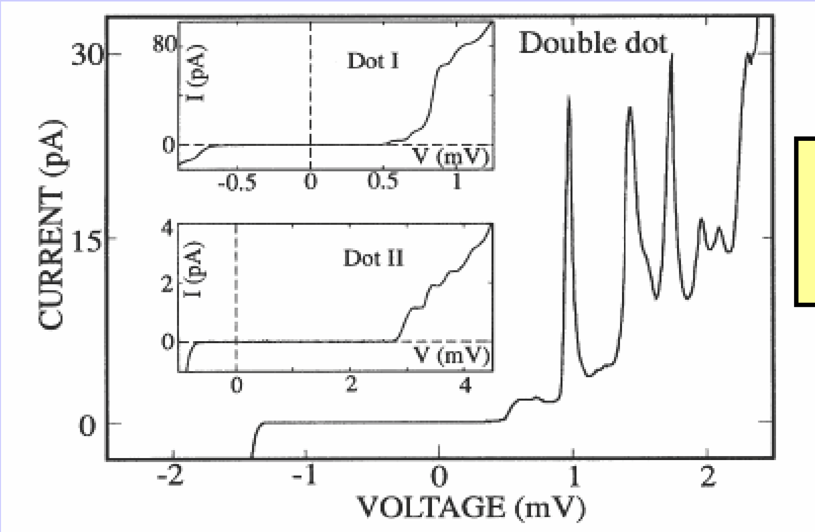
Questions that are Raised

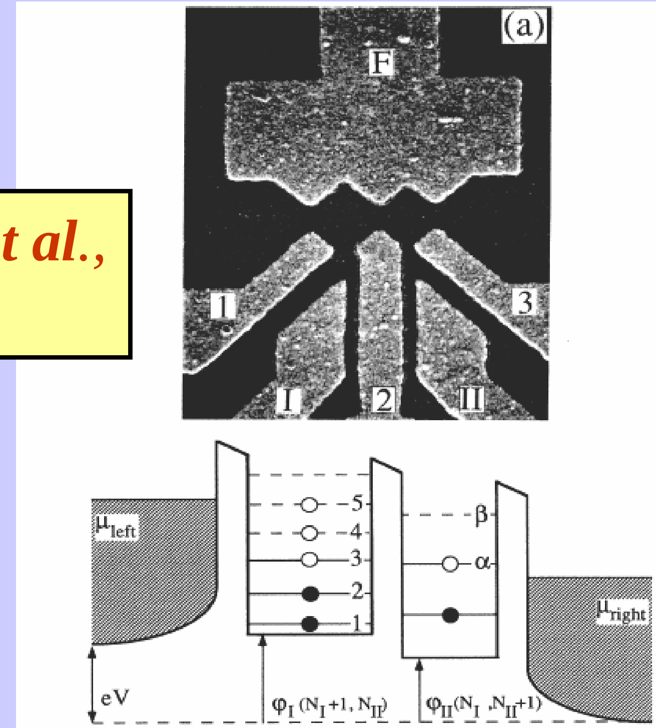
1. **Why peaks instead of steps?**

2. Why is the Coulomb blockade absent?

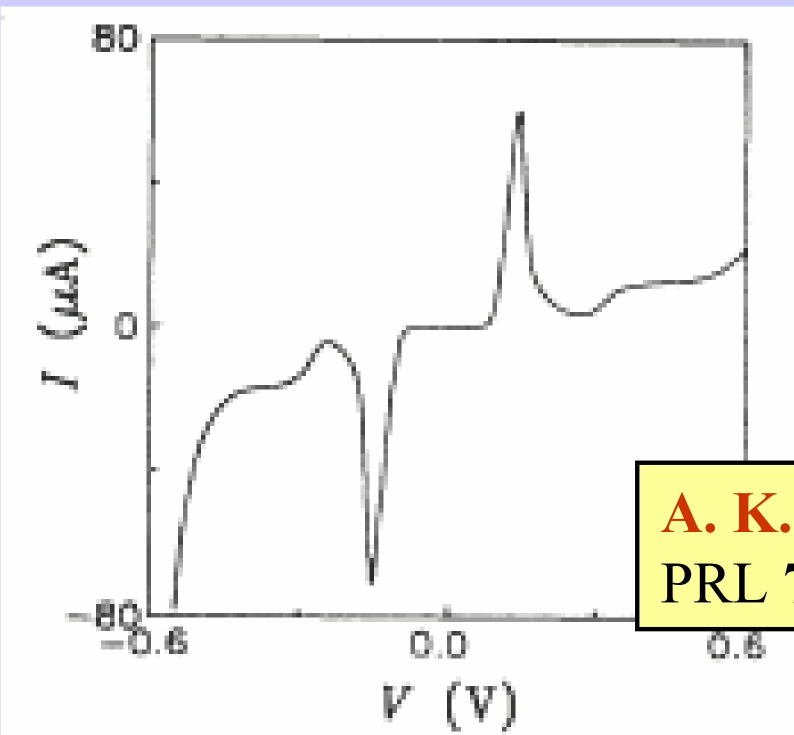
3. Why are the I(V) curves dependent on position?

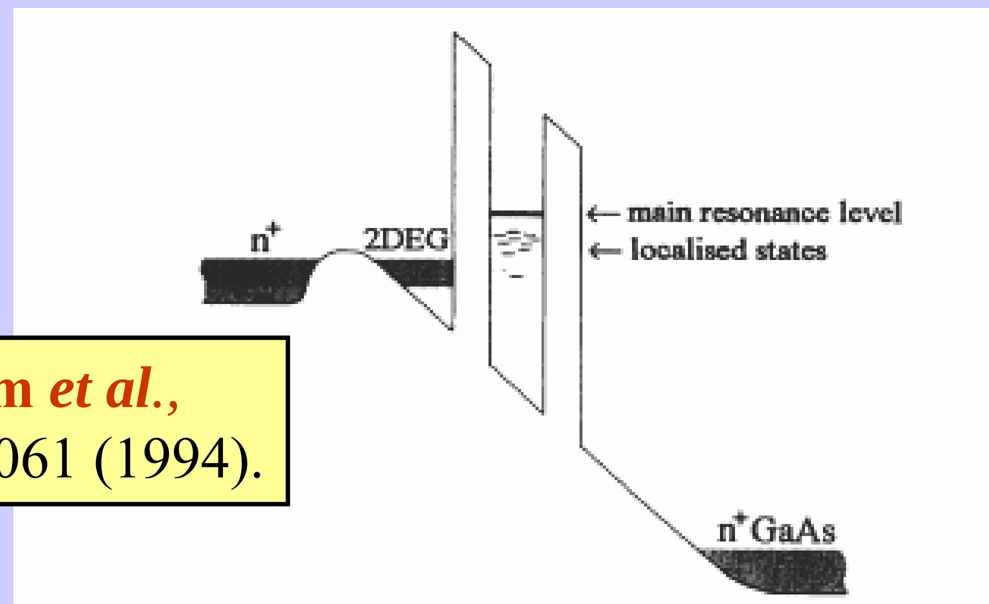
Peaks in the Current Voltage Curves: Resonant Tunneling Process





N. C. van der Vaart *et al.*,
PRL **74**, 4702 (1995).

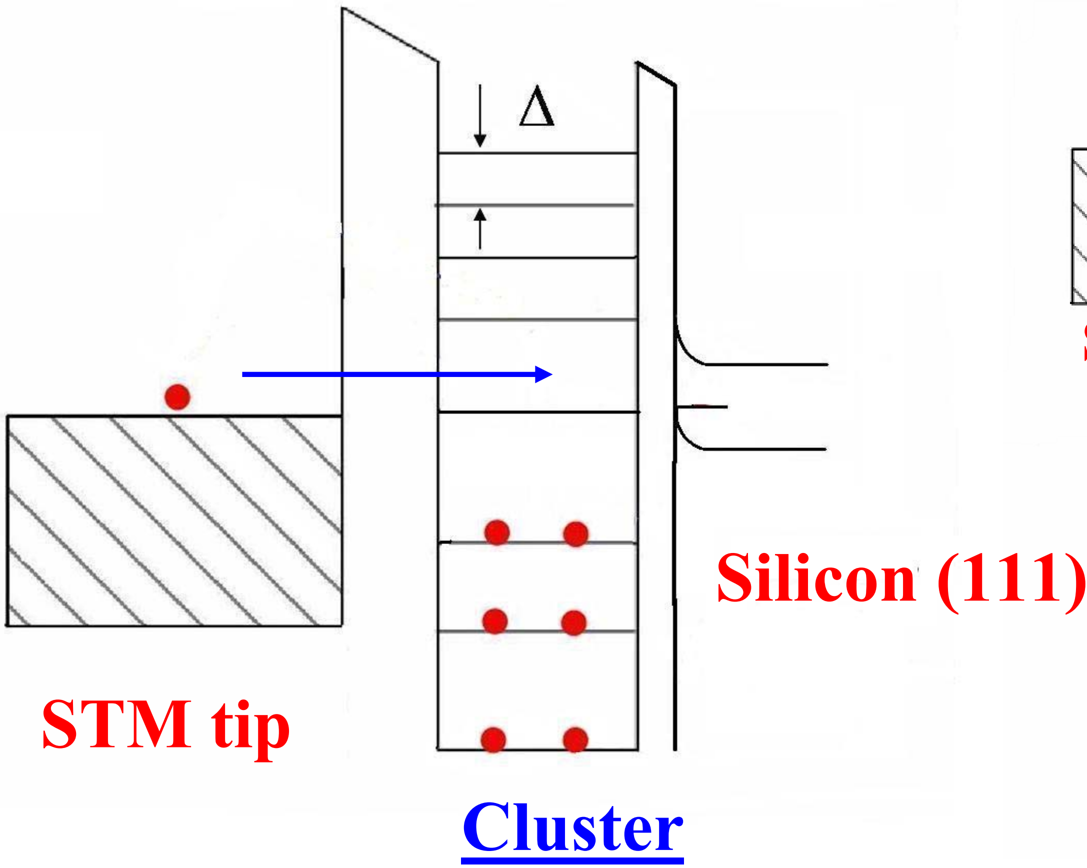




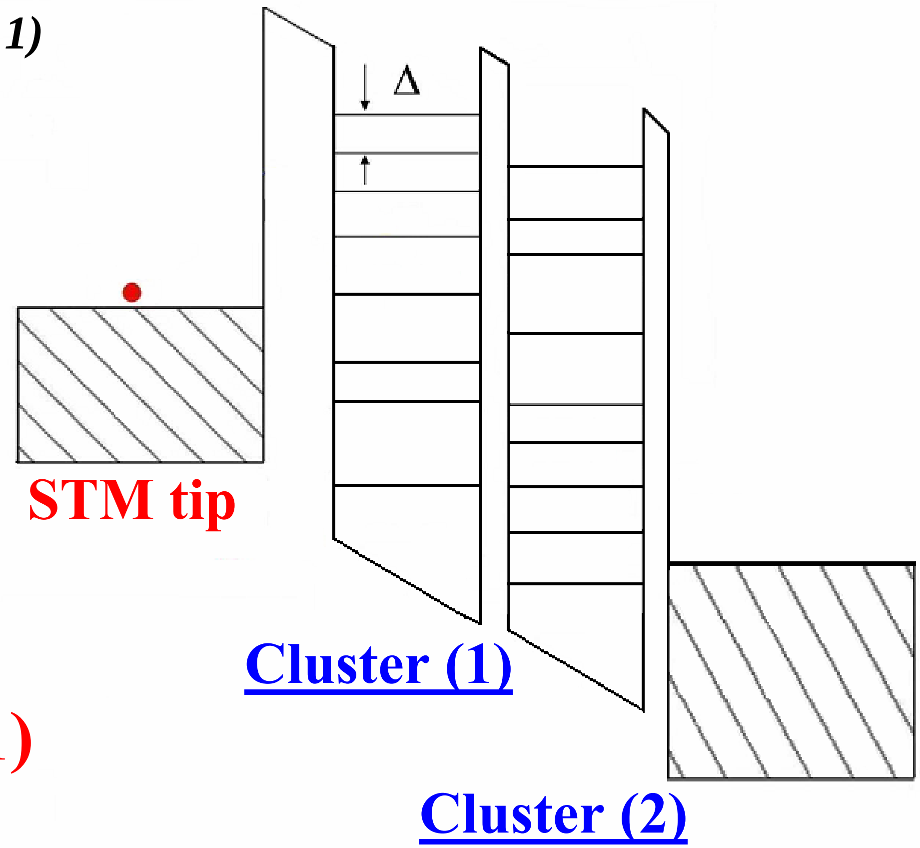
A. K. Geim *et al.*,
PRL **72**, 2061 (1994).

Peaks in the Current-Voltage Curves

There is an interface state between the Pb and Si(111) that can be conductive.



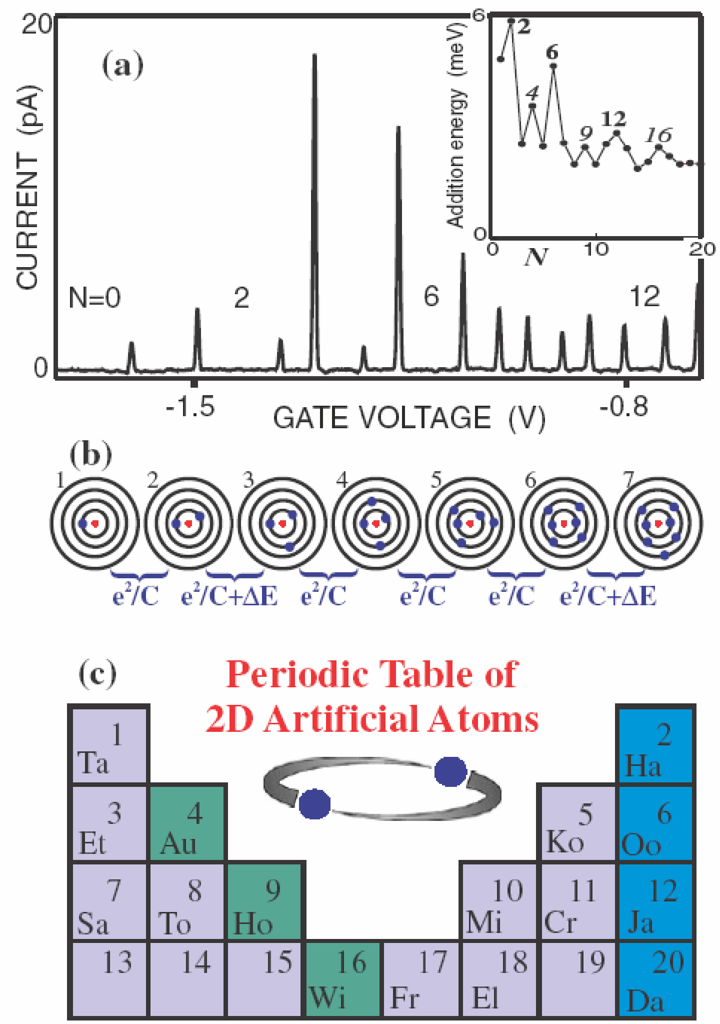
Case 1

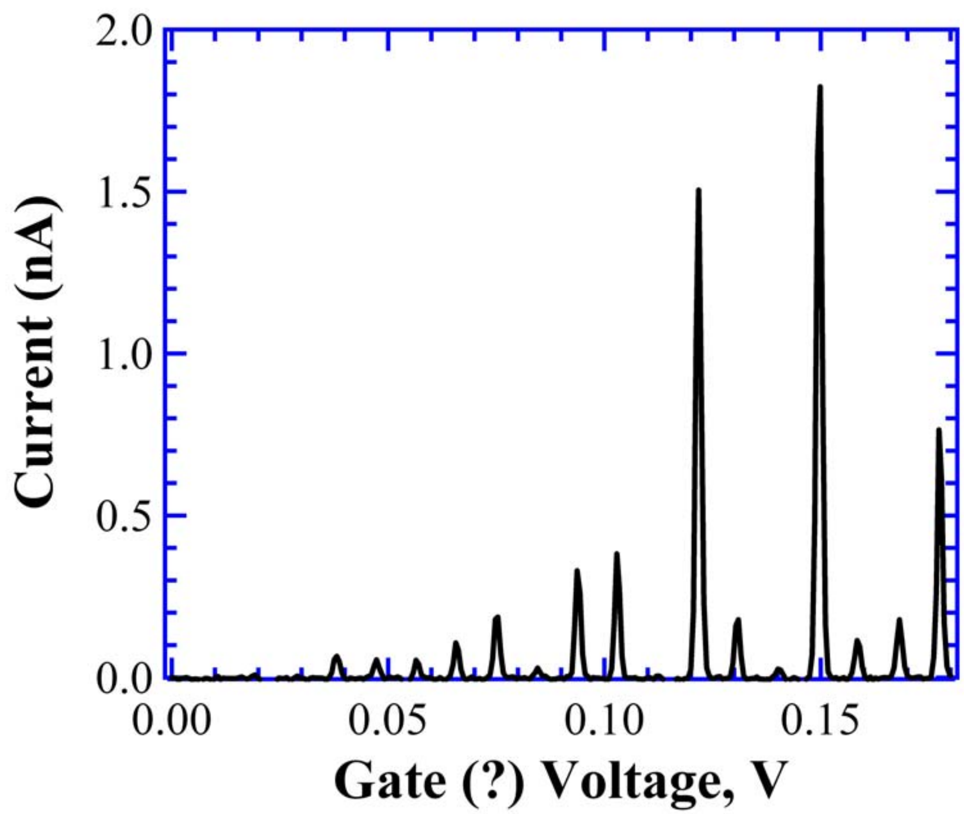


Case 2

Weitering, Ettema and Hibma,
PRB **45**,9126 (1992).

Peaks in the Current- (Gate ?) Voltage





L. P. Kouwenhoven, D. G. Austing, and S. Tarucha, Rep. Prog. Phys. 64, 701 (2001).

Questions that are Raised

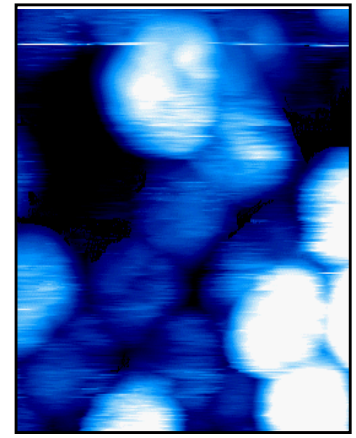
1. Why peaks instead of staircases?

2. **Why is the Coulomb blockade absent?**

3. Why are the I(V) curves dependent on position?

Absence of Coulomb Blockade and Charging Energy

The **capacitance** is much **larger** than estimates which are based exclusively on cluster size because:



1.) the nearby clusters add additional capacitive terms.

2.) the capacitance between the **tip and cluster** and **cluster and substrate** add additional terms.

Both of these factors would increase the capacitance and **decrease** the **charging energy**.

Questions that are Raised

1. Why peaks instead of staircases?

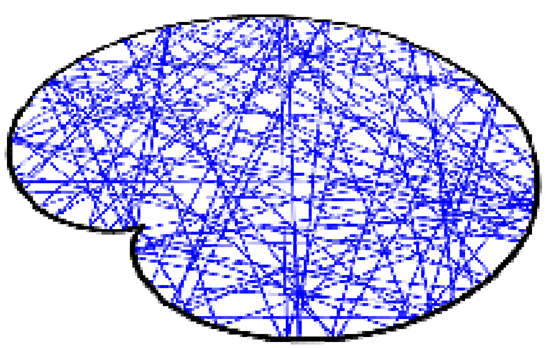
2. Why is the Coulomb blockade absent?

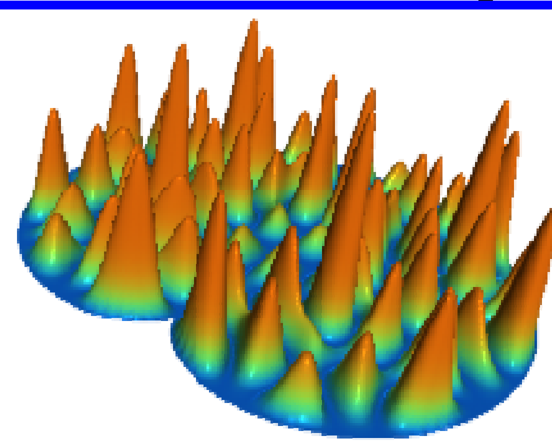
3. **Why are the I(V) curves dependent on position?**

Position Dependent Current-Voltage Curves

The position dependence is reminiscent of **Quantum Chaos**. The eigenstates are extended on the cluster but the eigenfunctions can be position dependent. Since the tunneling current is proportional to the tunneling matrix element squared (an expression relating the eigenfunctions), the variation in peak current amplitudes will be a function of position.

$$\boxed{\mathbf{M} = \hbar/2m\int (\Psi^*_{tip}\nabla\Psi_s - \Psi^*_s\nabla\Psi_{tip})\, dS}$$





From Arnd Baecker's homepage

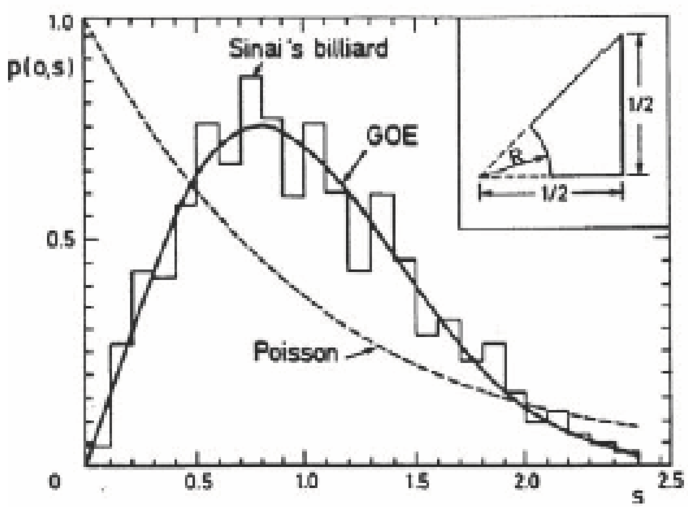
Bohigas - Giannoni - Schmit Conjecture

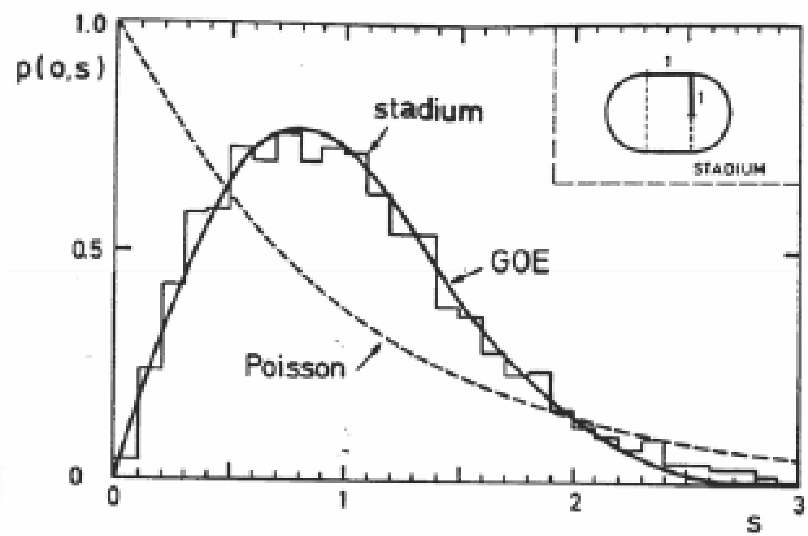
Characterization of Chaotic Quantum Spectra and Universality of Level Fluctuation Laws

O. Bohigas, M. J. Giannoni, and C. Schmit
Division de Physique Théorique, Institut de Physique Nucléaire, F-91406 Orsay Cedex, France
(Received 2 August 1983)

It is found that the level fluctuations of the quantum Sinai's billiard are consistent with the predictions of the Gaussian orthogonal ensemble of random matrices. This reinforces the belief that level fluctuation laws are universal.

In summary, the question at issue is to prove or disprove the following conjecture: Spectra of time-reversal-invariant systems whose classical analogs are K systems show the same fluctuation properties as predicted by GOE





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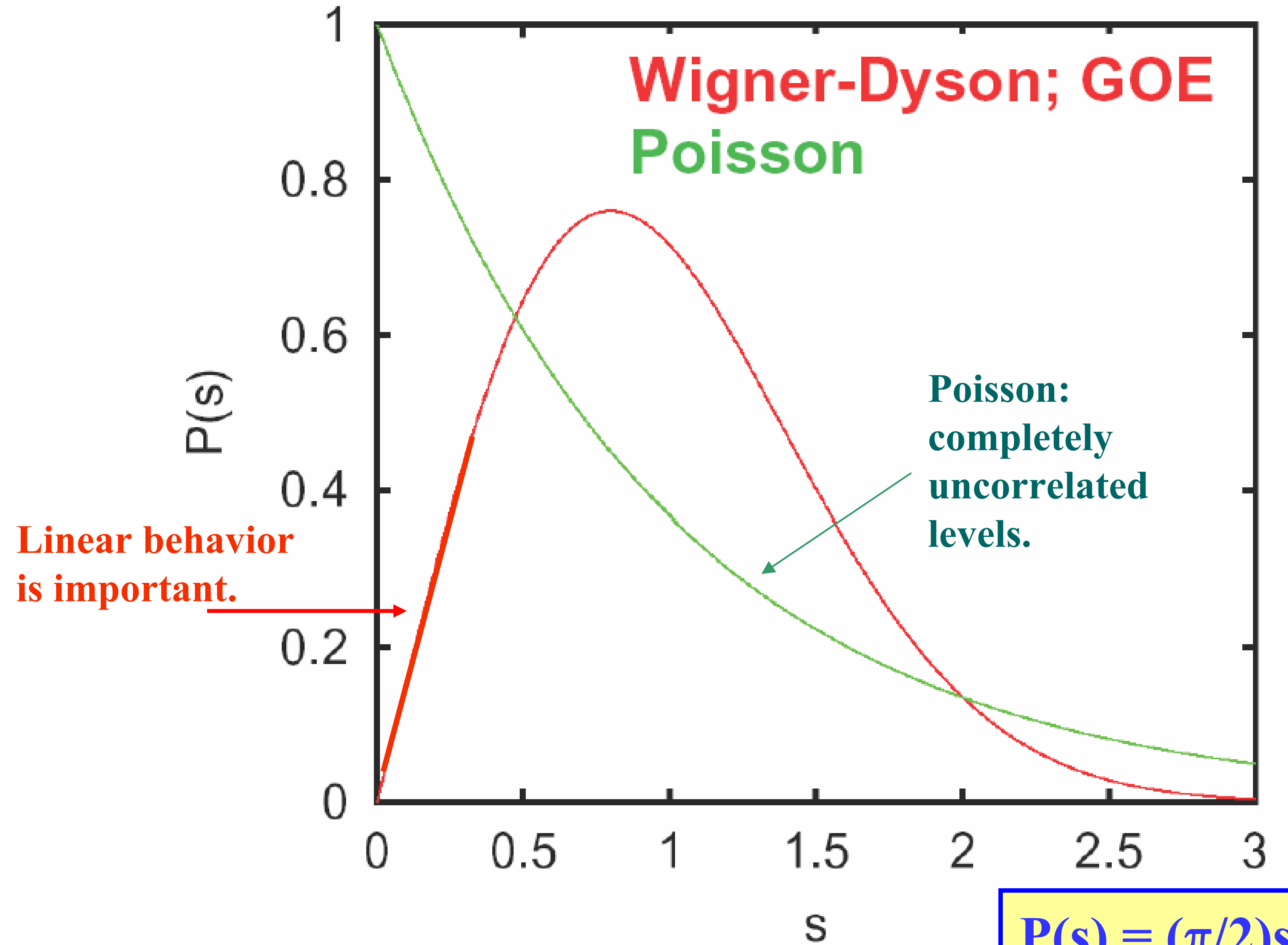
Statistical Distributions: Random Matrix Theory



GOE: Gaussian Orthogonal Ensemble

(real, symmetric Hamiltonian matrix.)

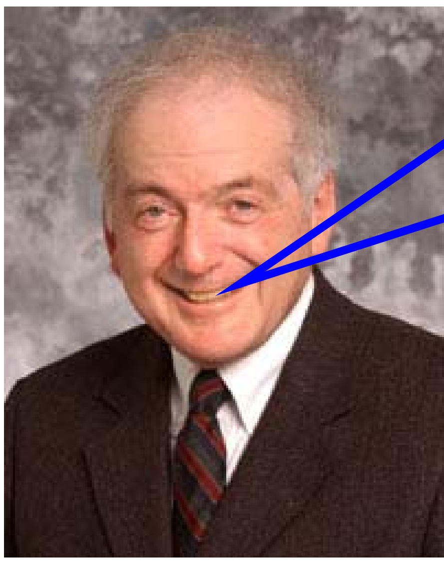
$$P(s) = (\pi/2)s \exp(-\pi s^2/4)$$

Plot from Professor Altshuler's talk: *"Introduction to Mesoscopics"*
http://www.lancs.ac.uk/users/esqn/windsor04/handouts/altshuler.pdf

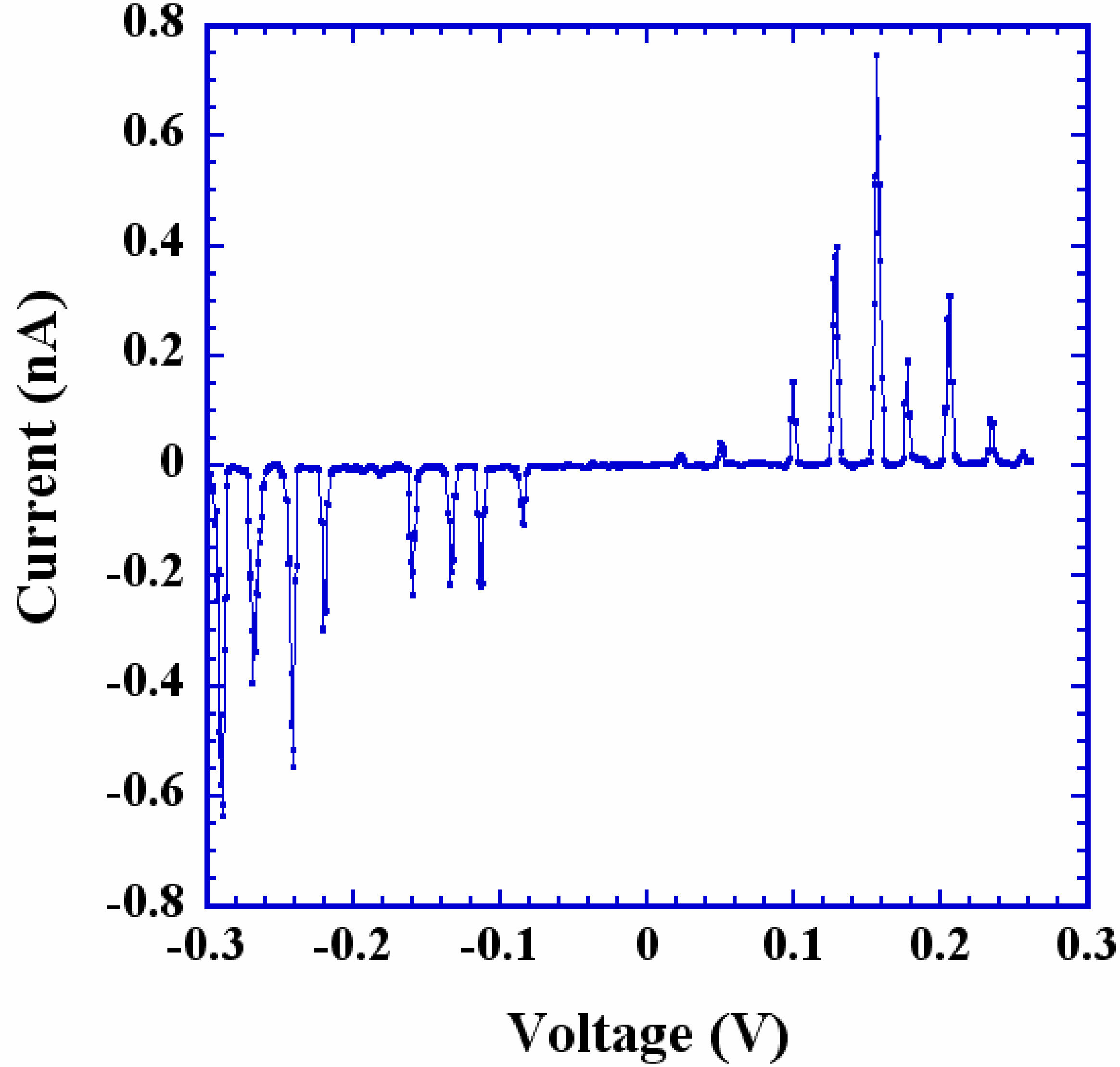
Interesting aspects of the STM data

Unequal and Irregular Peak Spacings

Unequal and Irregular Peak Heights



Study the spacings between the peaks.



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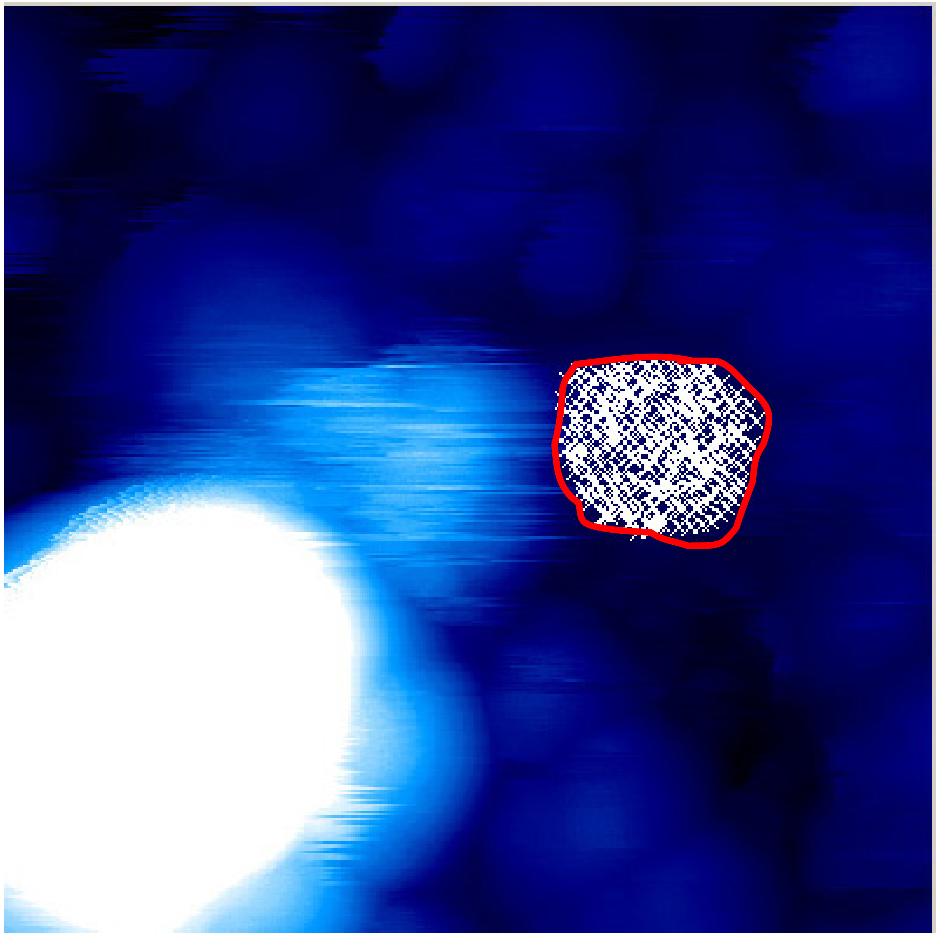
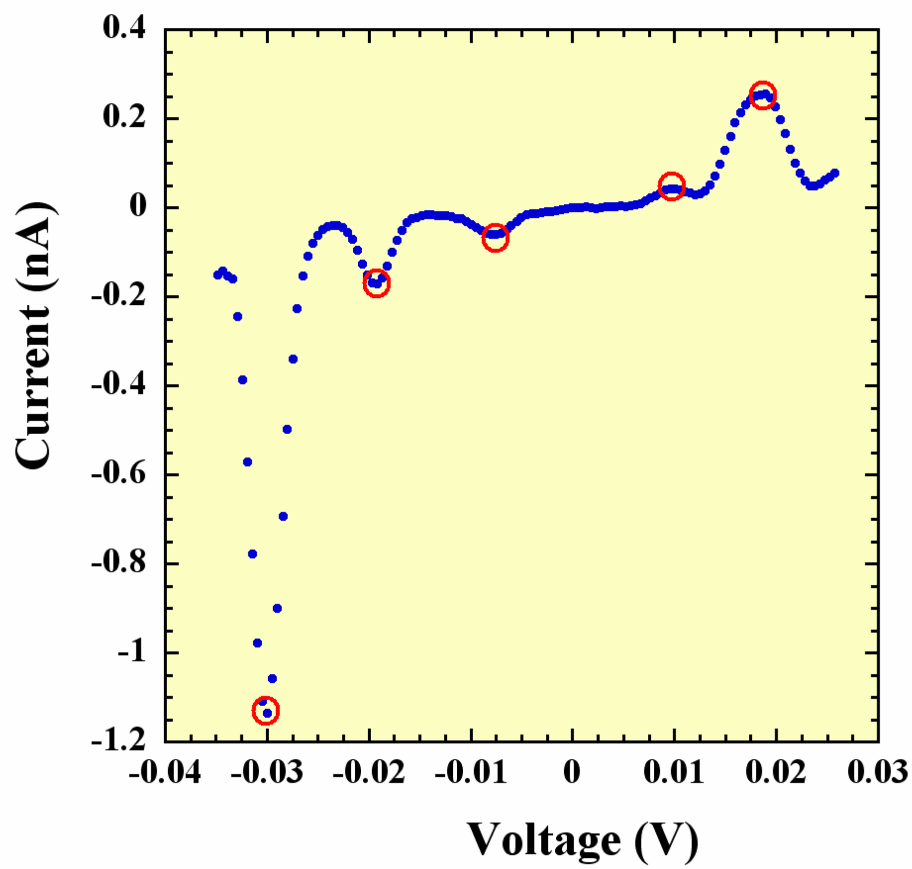
"280" I(V) Measurements through a Cluster



20 nm x 20 nm

Cluster Size:
Diameter ~ 44 Å
Height ~ 6 Å

Typical I(V) curve used in the analysis of the distributions



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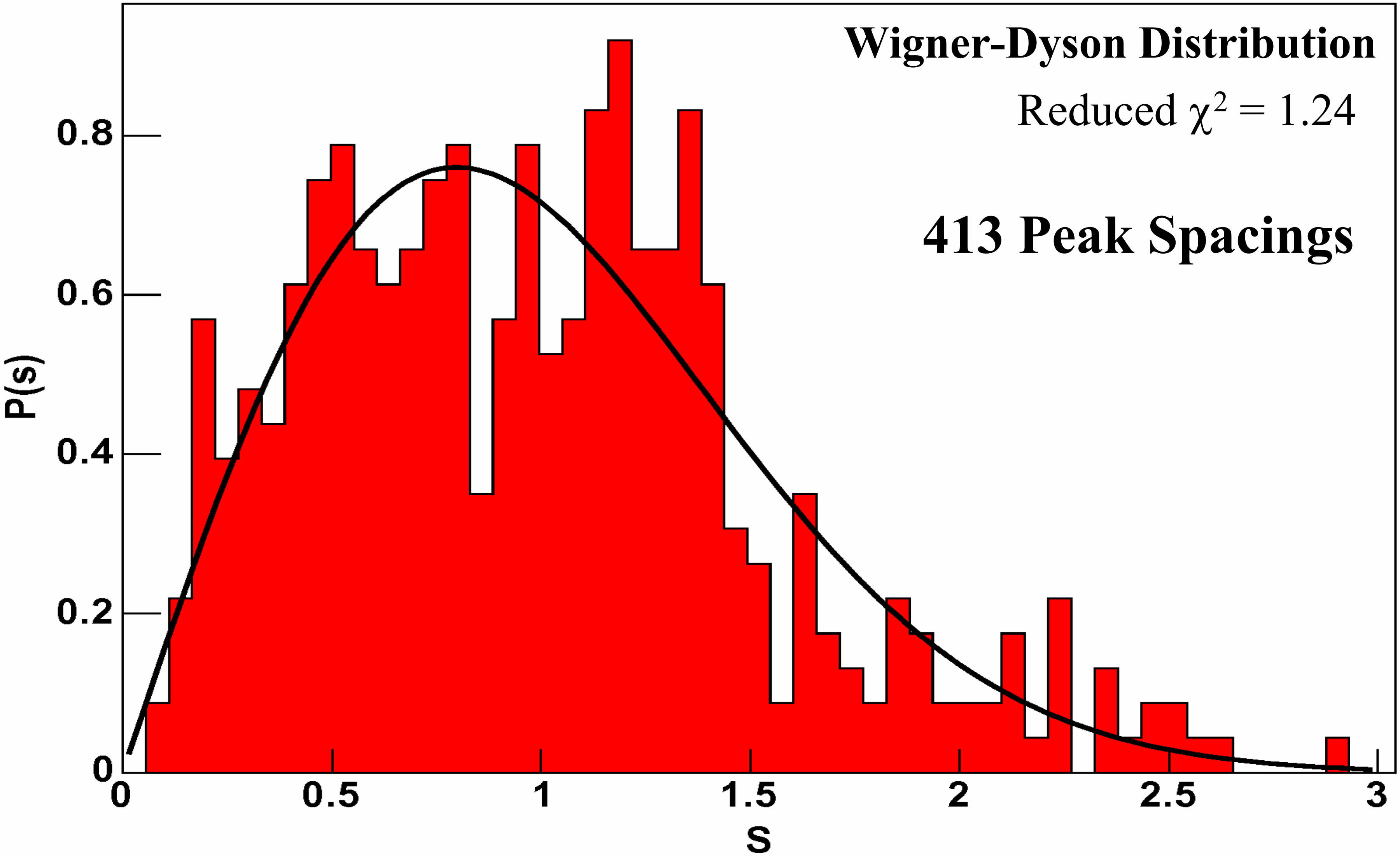
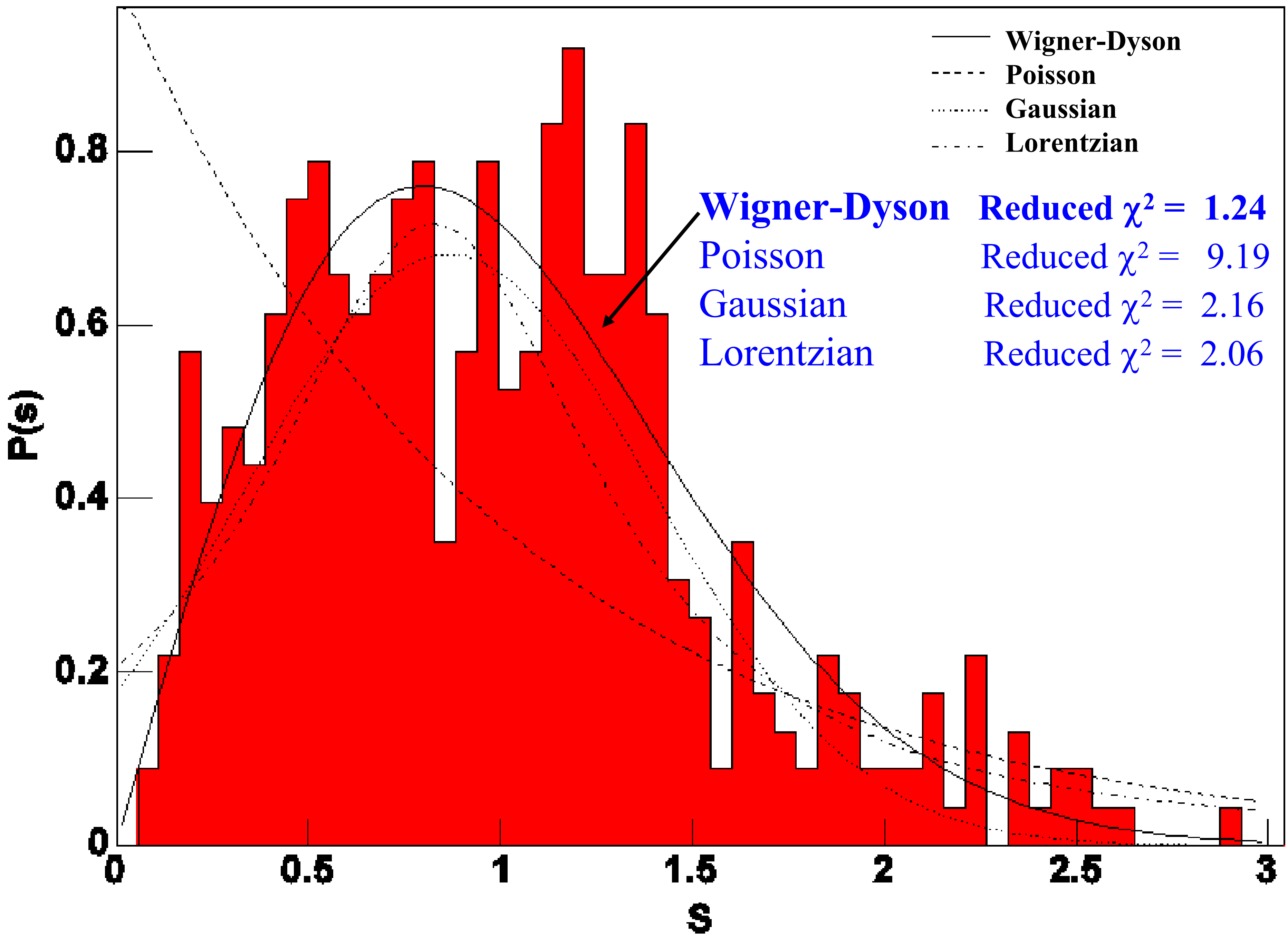
Peak Spacing Distribution

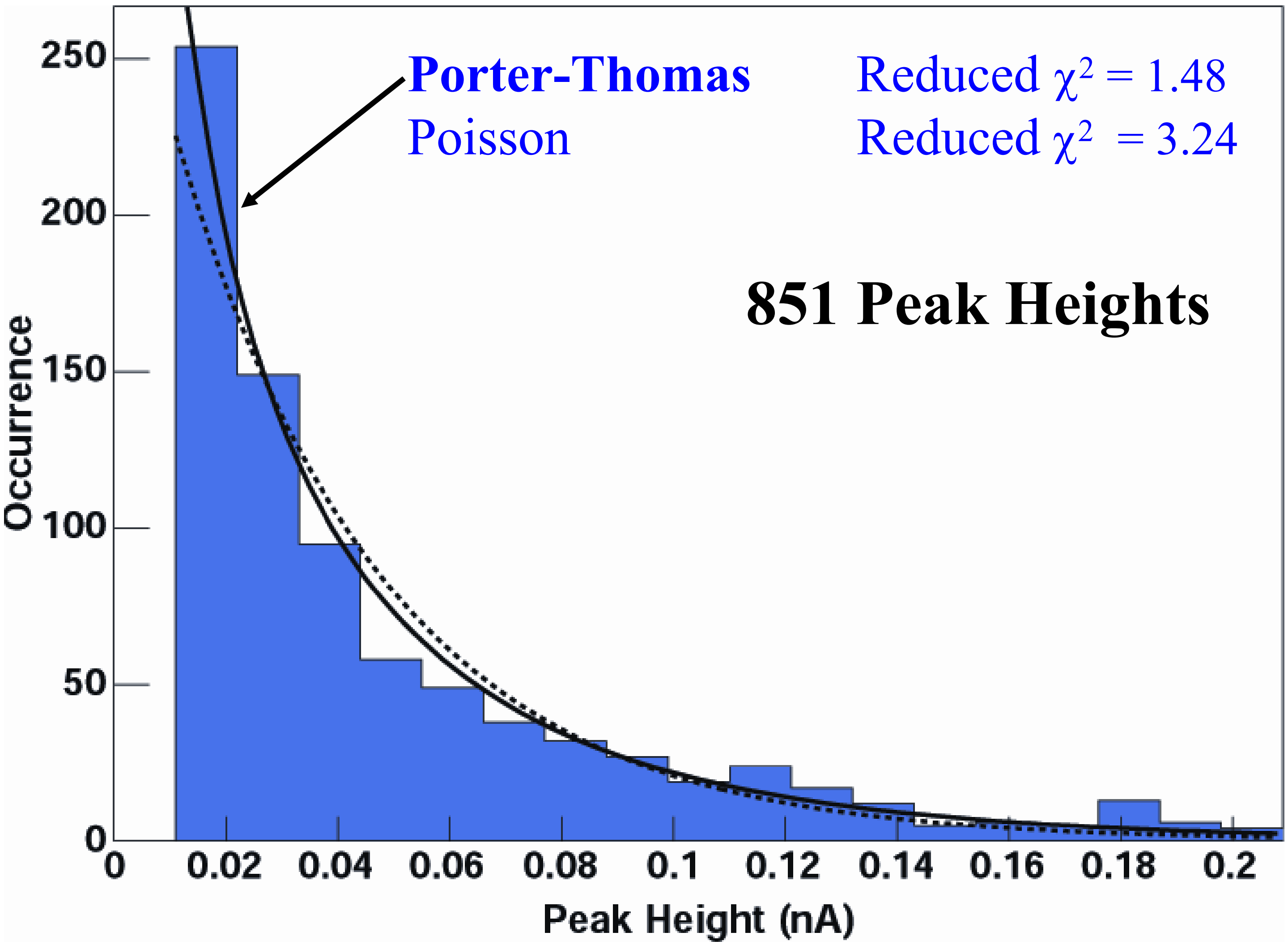


$$P(s) = (\pi/2)s \exp(-\pi s^2/4)$$

Comparing Different Distributions



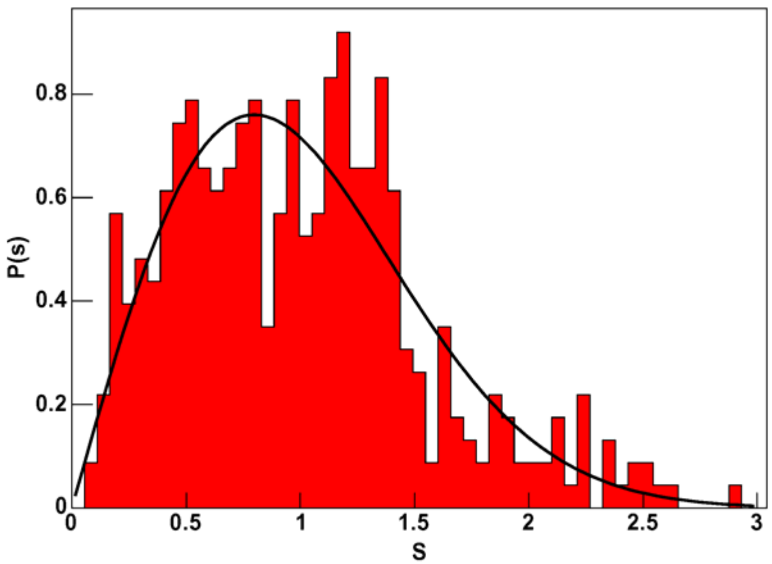
Peak Height Distribution



$$P(I) = (2\pi)^{-(1/2)} (I/\langle I\rangle)^{-(1/2)} \exp[-I/(2\langle I\rangle)]$$

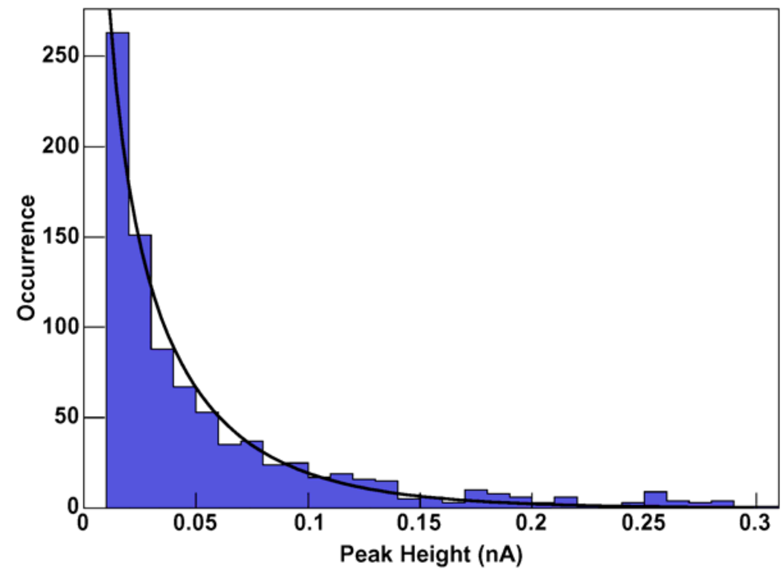
Experimental Evidence of RMT for both the Eigenvalues and the Eigenfunctions in a Metallic Cluster

Wigner-Dyson Distribution



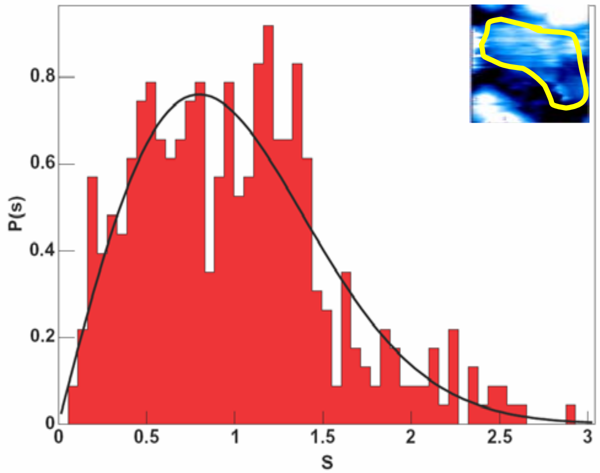
Eigenvalues

Porter-Thomas Distribution

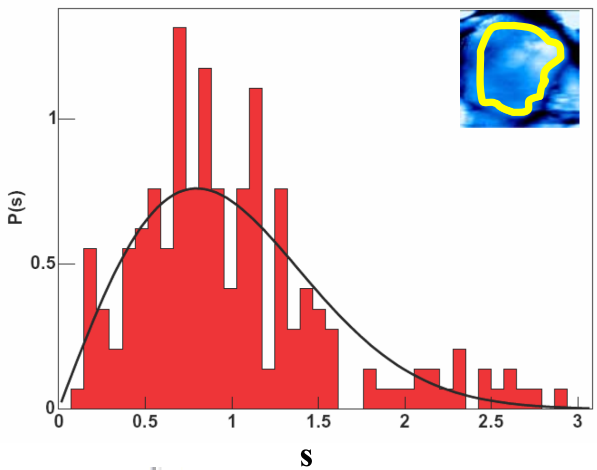


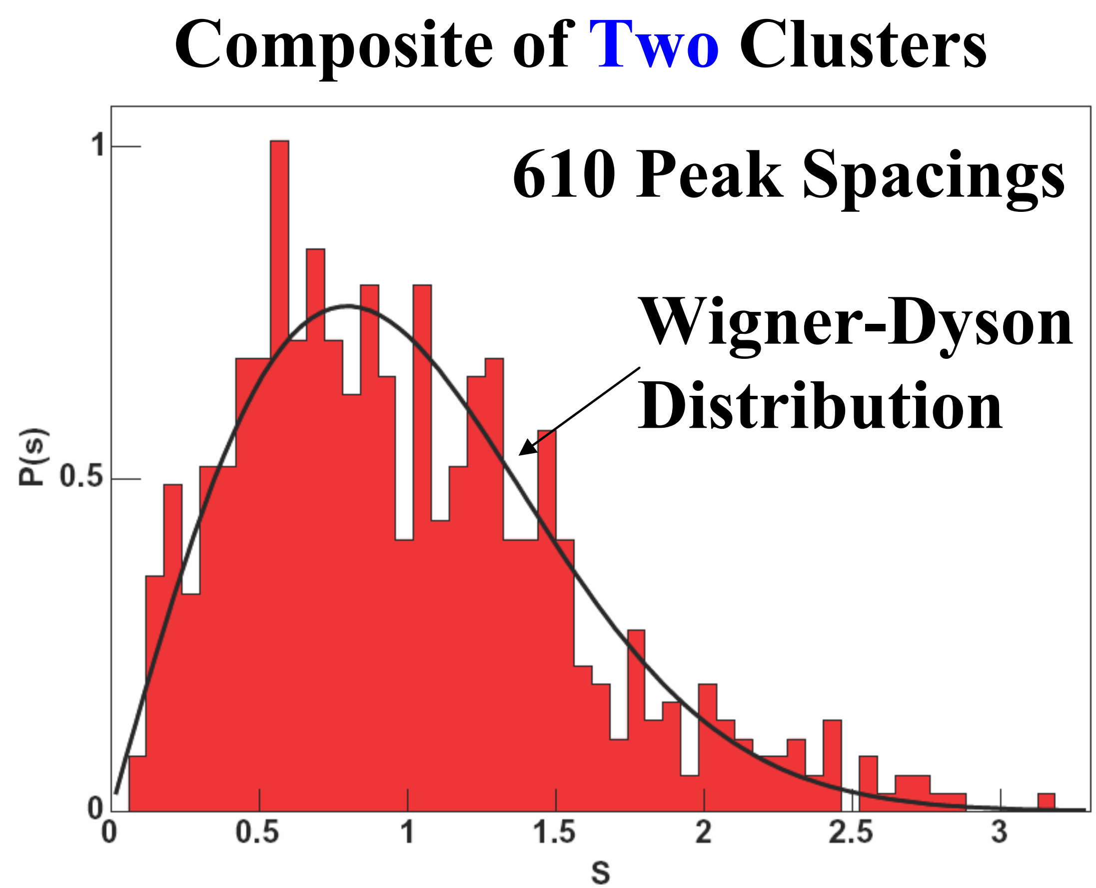
Eigenfunctions

Adding 2 Wigner Dyson Distributions



+

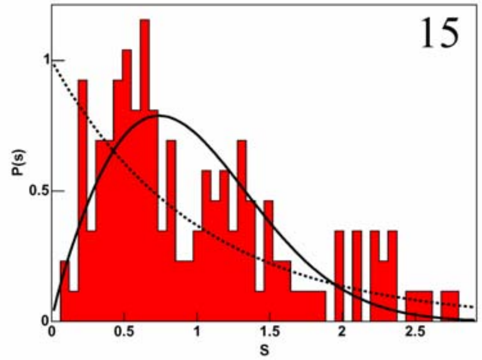




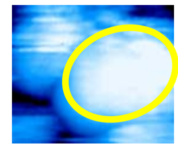
Transition to Quantum Chaos in Metallic Clusters

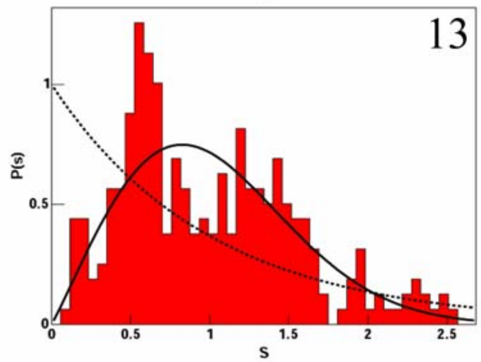
Poisson-like

Order

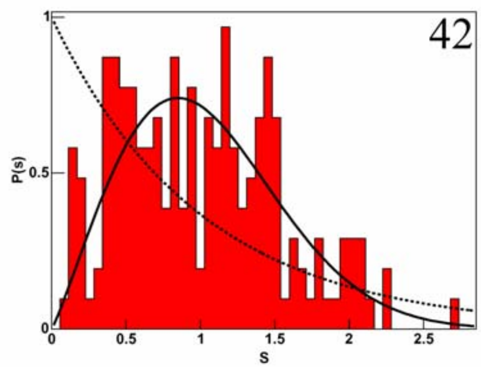


Volume ≈ 10 nm^3

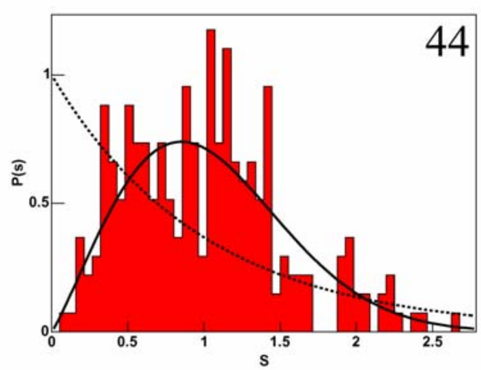




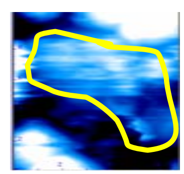
Volume ≈ 7.8 nm^3



Volume ≈ 9.6 nm^3



Volume ≈ 7.4 nm^3



Wigner-Dyson

Chaos

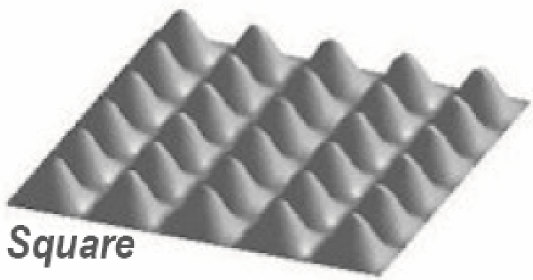
Imaging Quantum Chaos

VOLUME 85, NUMBER 11 PHYSICAL REVIEW LETTERS 11 SEPTEMBER 2000

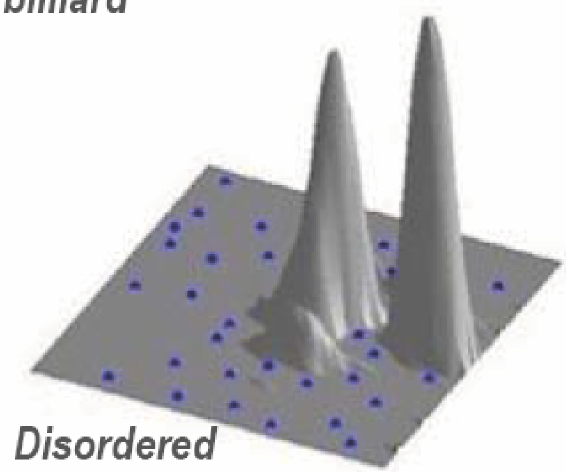
Correlations due to Localization in Quantum Eigenfunctions of Disordered Microwave Cavities

Prabhakar Pradhan and S. Sridhar
Department of Physics, Northeastern University, Boston, Massachusetts 02115
(Received 28 February 2000)

Integrable



Square billiard

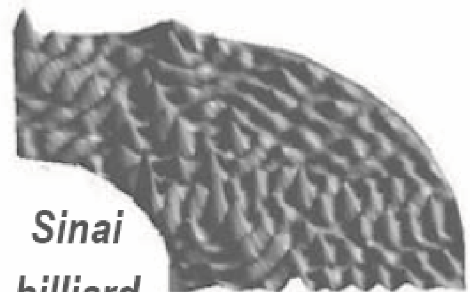


Disordered localized

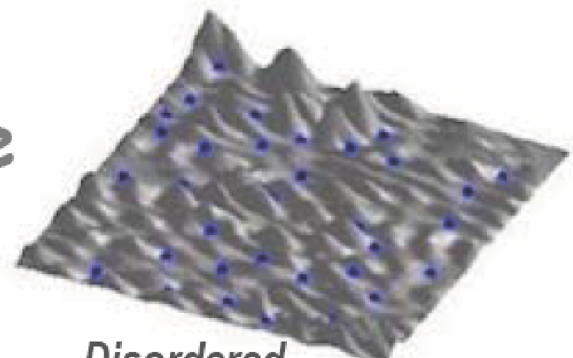
All chaotic systems resemble each other.

All integrable systems are integrable in their own way

Chaotic



Sinai billiard

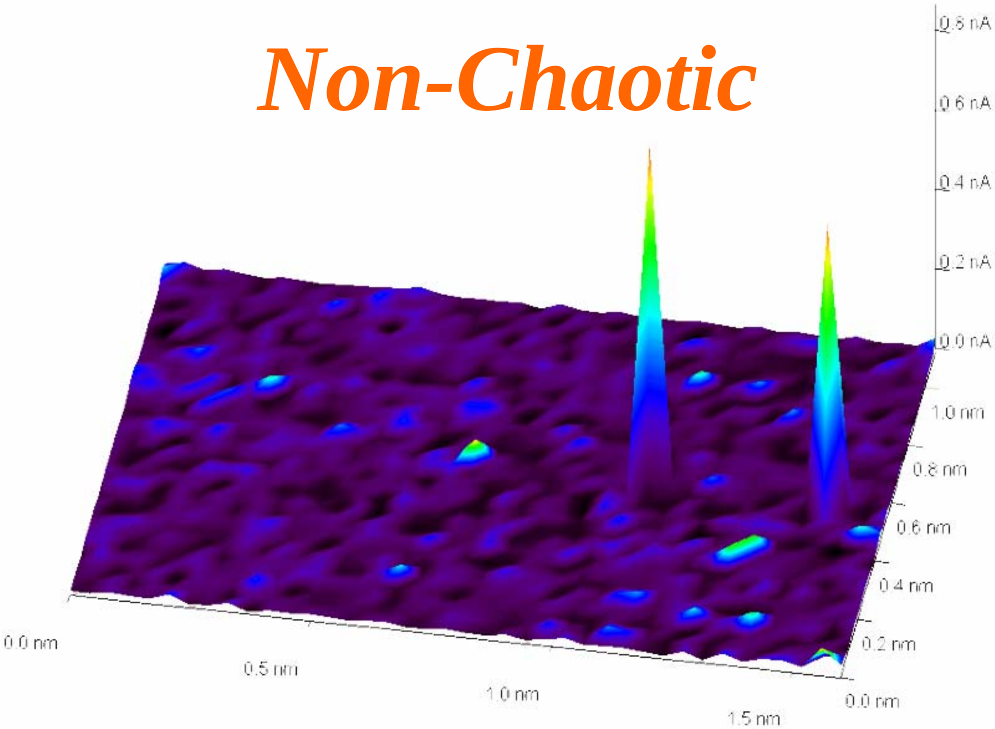


Disordered extended

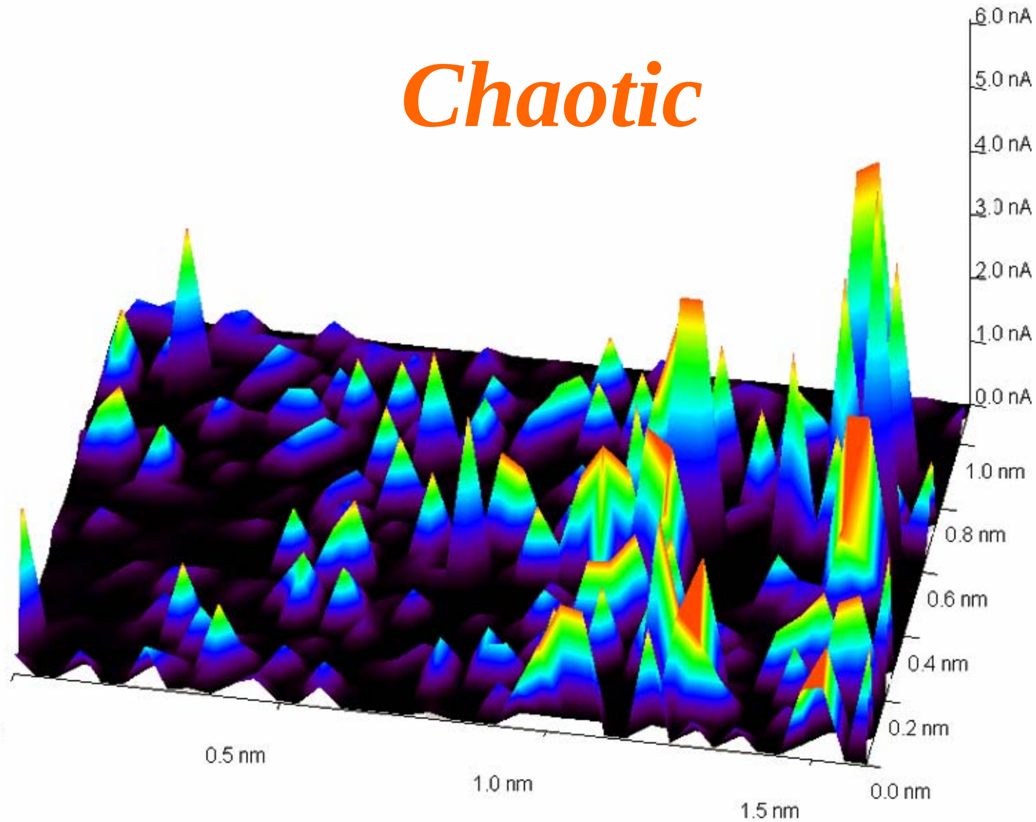
Plot from Professor Altshuler's talk: *"Introduction to Mesoscopics"*
http://www.lancs.ac.uk/users/esqn/windsor04/handouts/altshuler.pdf

Imaging "Quantum Chaos" with an STM

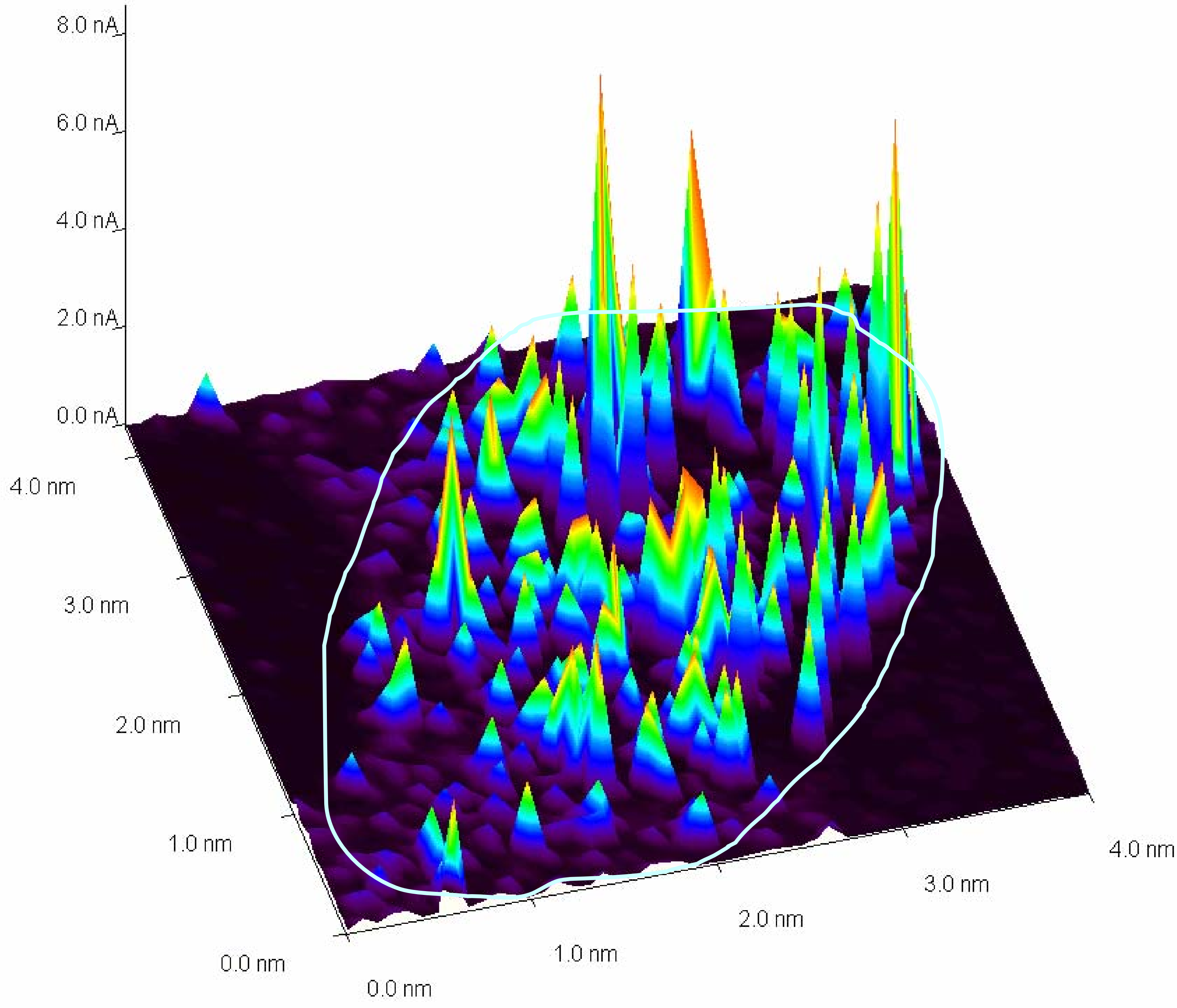
Non-Chaotic



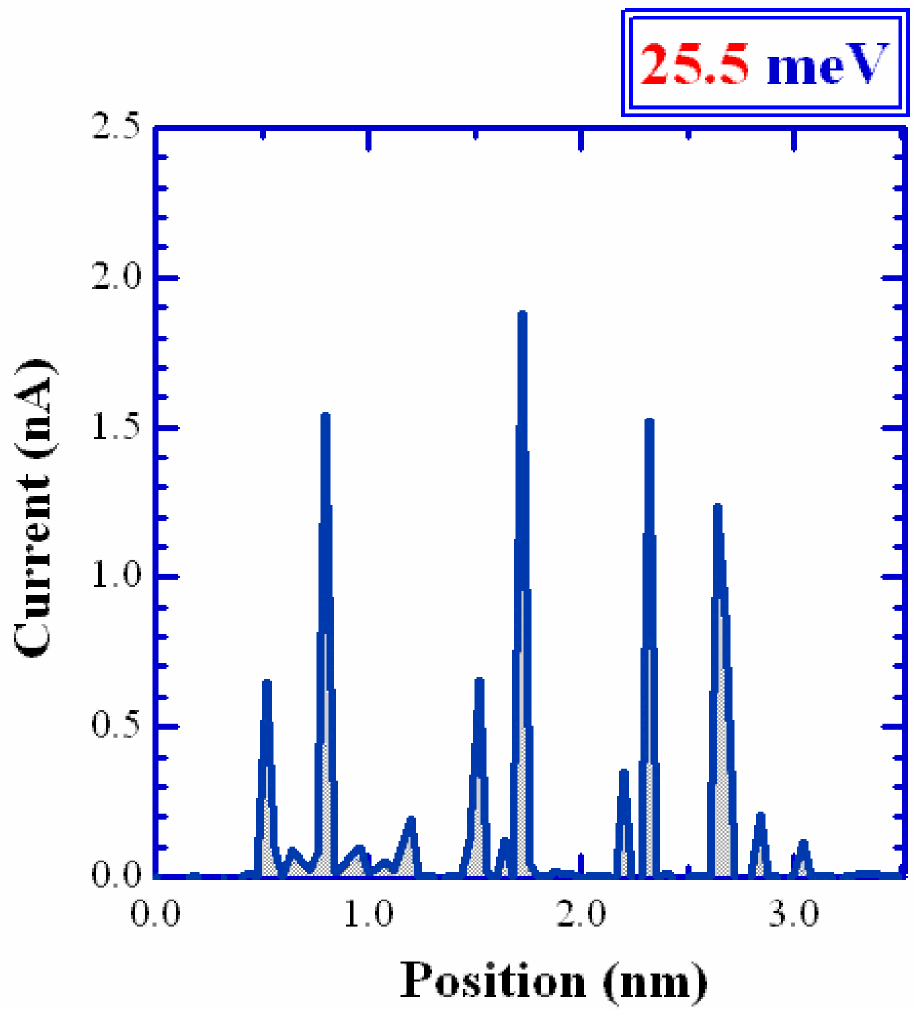
Chaotic



"Quantum Chaos" in a Metallic Cluster



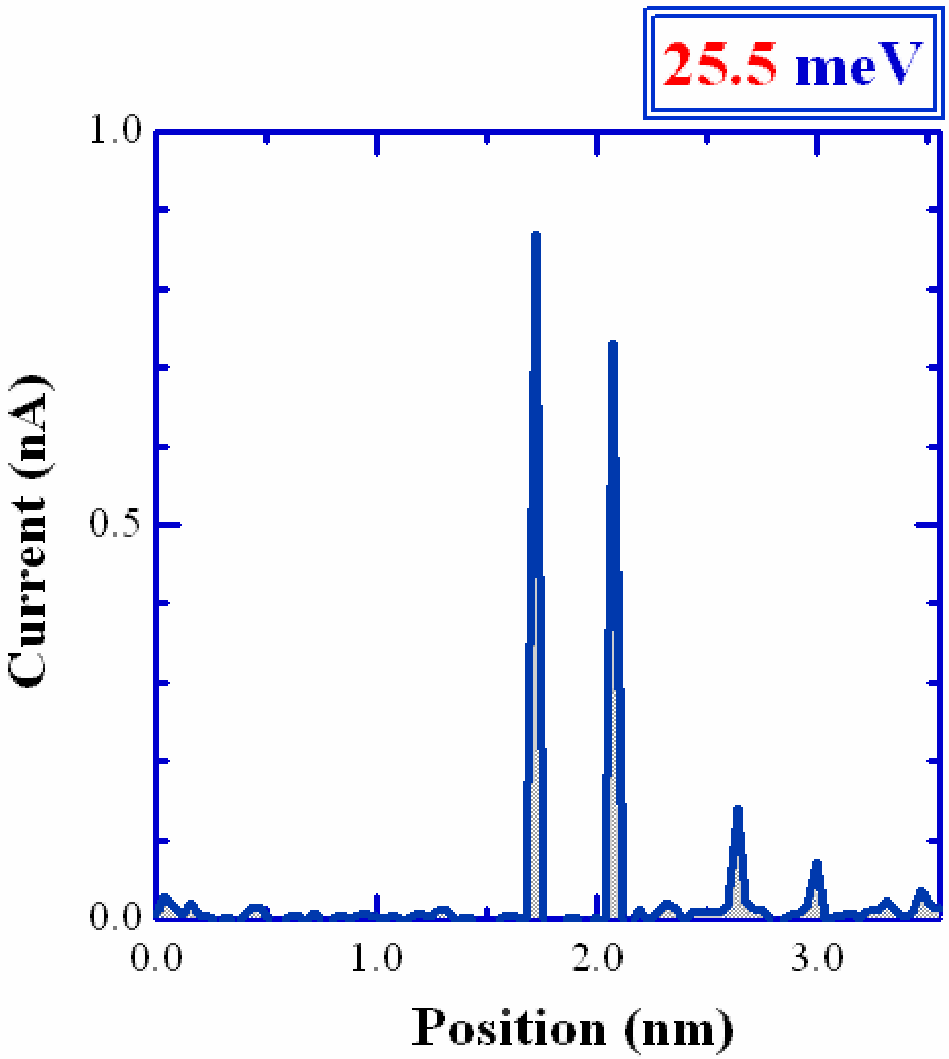
Current as a Function of Position Along a “Chaotic” Cluster



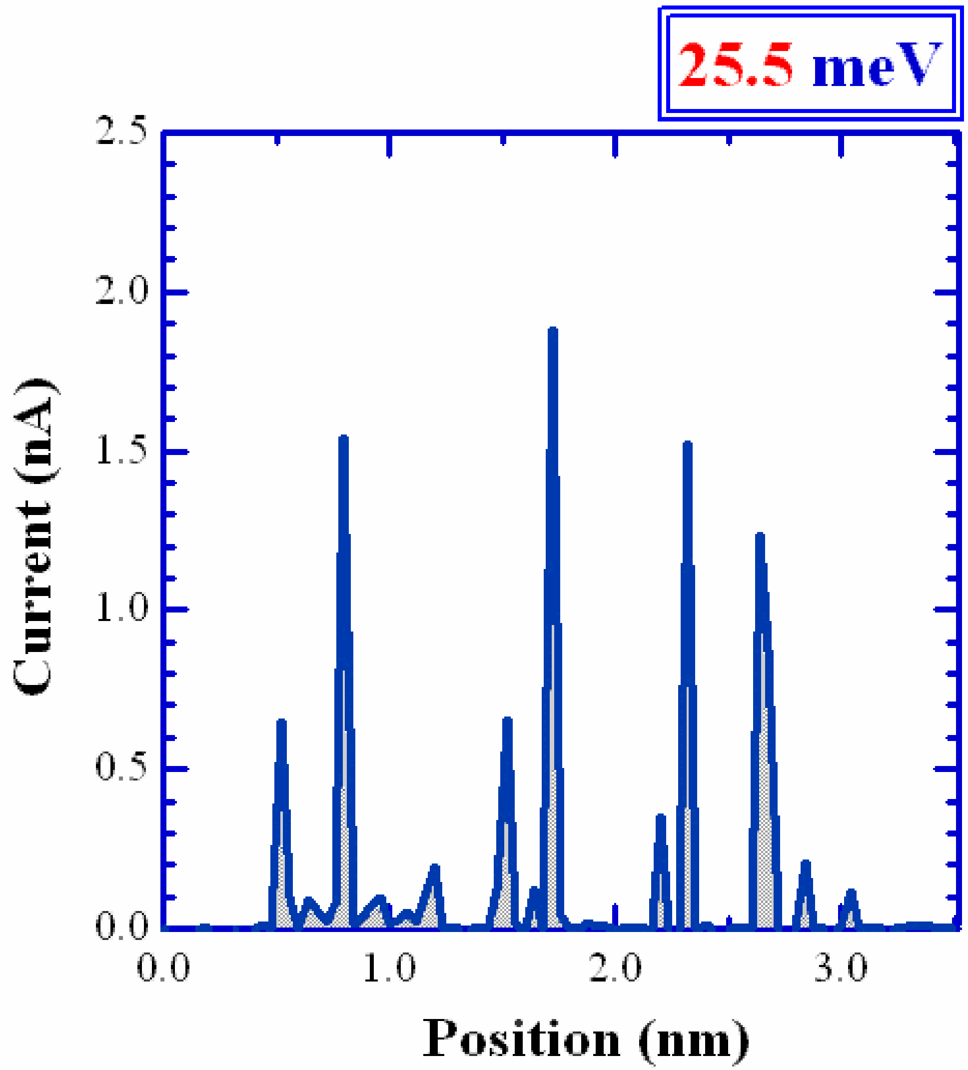
UNIVERSITY OF MINNESOTA

Current as a Function of Position Along a Cluster

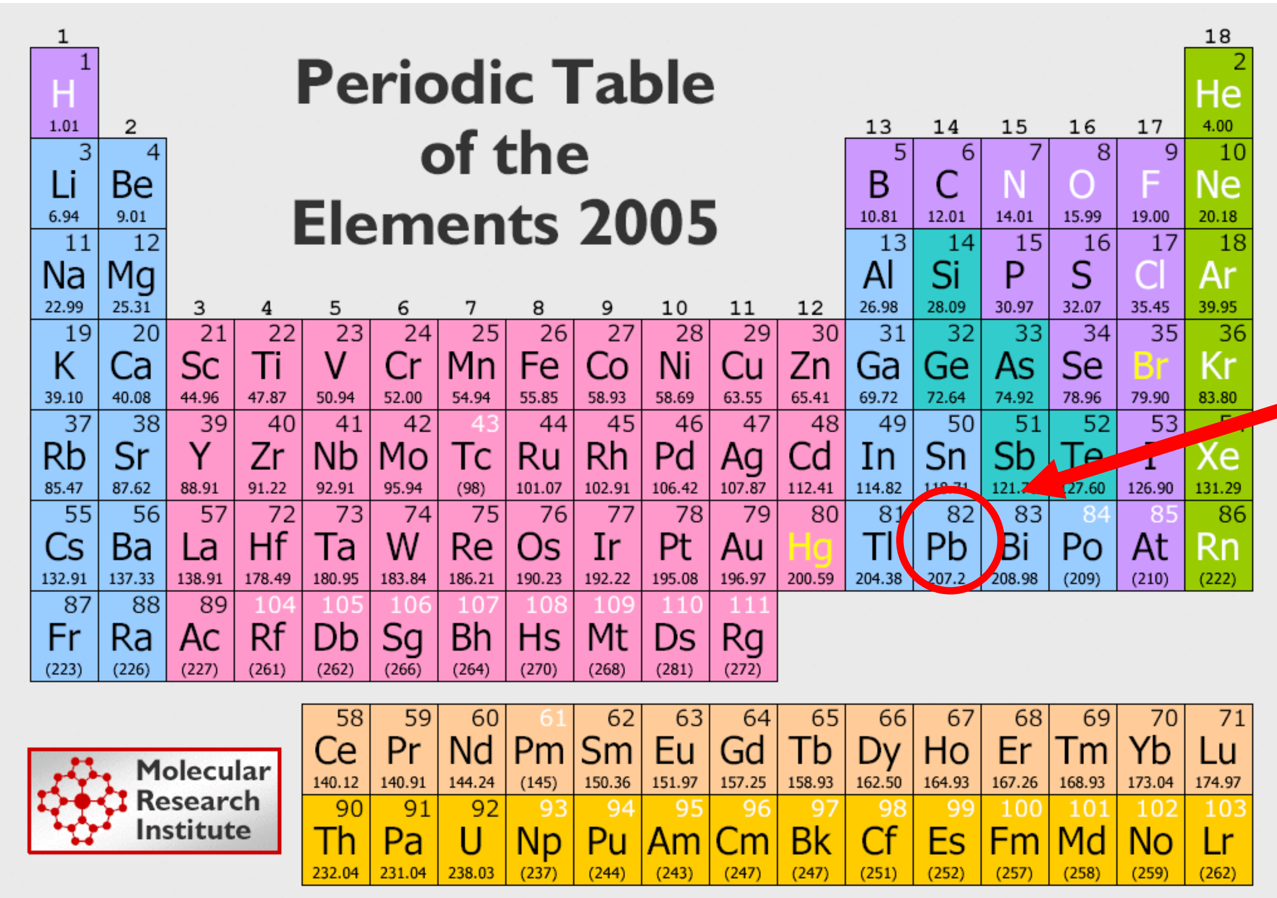
Non-Chaotic



Chaotic



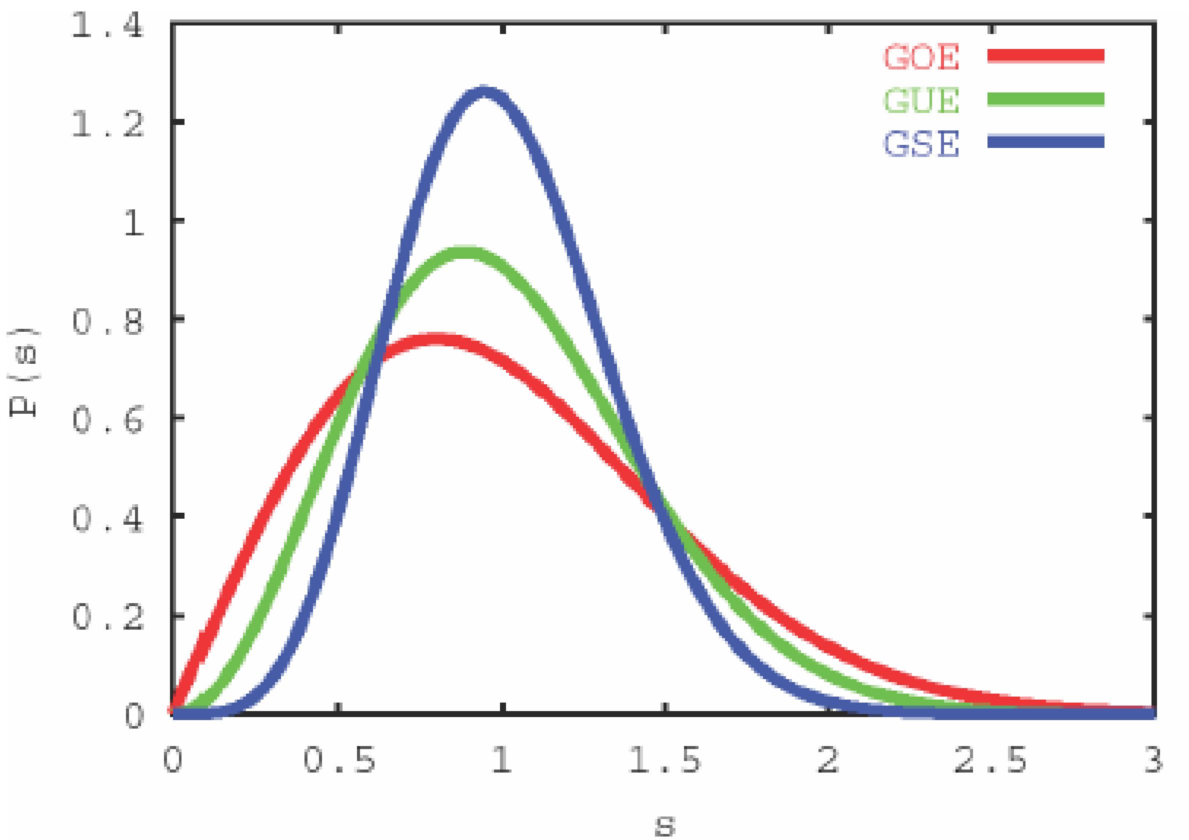
Possible "Fly in the Ointment": Spin-Orbit Interactions



- High Z material.
- Pb Z = 82
- Spin-Orbit interactions should be strong for a high Z material.

Spin-Orbit Interactions

- High Z material. Pb Z = 82
- The statistics should follow GSE (Gaussian Symplectic Ensemble) not Wigner-Dyson (GOE).
- However, we don't see GSE in our distributions.



Suppression of Spin-Orbit Scattering in Au Grains

Suppression of Spin-Orbit Scattering in Strongly Disordered Gold Nanojunctions

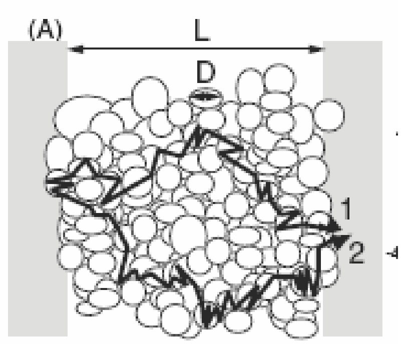
A. Anaya, M. Bowman, and and D. Davidović

Georgia Institute of Technology, Atlanta, Georgia 30332, USA

(Received 4 February 2004; published 8 December 2004)

We discovered that spin-orbit scattering in strongly disordered gold nanojunctions is strongly suppressed relative to that in weakly disordered gold thin films. This property is unusual because in weakly disordered films spin-orbit scattering increases with disorder. Granularity and freezing of spin-orbit scattering inside the grains explains the suppression of spin-orbit scattering. We propose a generalized Elliot-Yafet relation that applies to a strongly disordered granular regime.

DOI: 10.1103/PhysRevLett.93.246604 PACS numbers: 72.25.Rb, 72.25.Ba, 73.21.La, 73.23.–b



Granular Au has a reduced spin-orbit scattering in comparison to Au films.

Open Questions

- Why are the spin-orbit interactions suppressed?
  Why not GSE statistics instead of GOE statistics?

- What is the nature of the tunneling process which results in current peaks as a function of bias voltage?

- Why does the Porter-Thomas distribution seem to describe the *current* amplitudes?

- Are nearby clusters acting as gates?

- Are there electrostatic effects which perturb the energy levels in the cluster as the bias voltage is changed?

Future Research Possibilities

- Apply a magnetic field.
  (GOE → GUE statistics)
- Use an oxidized metallic substrate and look at current steps as a function of distance along the cluster.
- Change the spacing between the clusters.
  (no Pt electrode)
- Reconstruct Si(111) and then grow clusters.
- Grow ferromagnetic clusters and study GSE.
- Measure temperature dependence.
- Apply a gate voltage.

Conclusions

Spectroscopic measurements resulted in current peaks which are position dependent on a cluster.

Peak spacings and peak heights are apparently consistent with expectations of Random Matrix Theory applied to a classically chaotic confined system.

There is an apparent transition in the mean level spacings as the cluster changes shape.

It is possible to use scanning tunneling spectroscopy to map out the square of eigenfunctions as a function of position.

<u>Physical Review Letters, 95, 146804</u>

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STM tip Fabrication

Ibrahim El-Sayed

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